RADIOECOLOGICAL SURVEILLANCE OF THE ENVIRONMENT

OBJECTIVE: TO ESTABLISH Pu AND Am CONCENTRATIONS LEVELS IN THE DIFFERENT COMPARTIMENTS OF THE ENVIRONMENT AND THEIR TIME EVOLUTION. TO ASSESS THE RADIOLOGICAL IMPACT ON THE POPULATION

ACTIVITIES:

* SAMPLING, ANALYSES AND MEASUREMENT OF:
  - SOILS
  - AIR
  - VEGETATION (WILD VEGETATION AND AGRICULTURAL PRODUCTS)
  - ANIMALS (INCLUDING DERIVED FOODS)

* DATA EVALUATION

* DOSE ASSESSMENT

RADIOECOLOGICAL RESEARCH

OBJETIVE: TO IMPROVE THE KNOWLEDGE ABOUT THE ENVIRONMENTAL BEHAVIOUR OF Pu AND Am IN THE AREA, LEADING TO BETTER UNDERSTANDING OF THE PROCESSES INVOLVED IN THE TRANSFER OF ACTINIDES TO MAN AND PARAMETERS GOBERNING THEM

ACTIVITIES:

* GEOCHEMICAL STUDIES
  - GRAIN SIZE DISTRIBUTION
  - MINERALOGICAL ASSOCIATIONS
  - MIGRATION IN DEPTH
  - SPECIATION

* RESUSPENSION
  - DISTRIBUTION OF RESUSPENDED PARTICLES (SIZE AND HEIGHT)
  - DETERMINATION OF INHALED FRACTION
  - RESUSPENSION FACTORS (TIME EVOLUTION)
  - DUST LOAMDING AND MASS LOANDING FACTORS
  - MODELLING
  - REDISTRIBUTION OF THE CONTAMINATION

* TRANSFER FACTORS (SOIL TO PLANT)

* DERIVED INTERVENTION LEVELS

* MARINE RADIOECOLOGY
  - LAND-SEA TRANSPORT OF Pu AND Am
  - Pu AND Am INVENTORIES IN THE SEA SEDIMENT
  - WATER- SEDIMENT TRANSFER OF Pu AND Am
  - Pu AND Am GEOCHEMISTRY IN THE SEA SEDIMENTS

RADIOLOGICAL SURVEILLANCE OF THE POPULATION

OBJETIVE: TO ASSES INDIVIDUAL DOSES BY BIOASSAY DATA AND DIRECT MEASUREMENT (WHOLE BODY COUNTER)

ASSUMPTION: INHALATION AS MAJOR CRITICAL PATHWAY

ACTIVITIES:

* URINE ANALYSES OF Pu AND Am (ALFA SPECTROMETRY MEASUREMENTS), 150 PERSONS BY YEAR

* WHOLE BODY COUNTER MEASUREMENTS, 20 PERSONS BY YEAR

* DOSE CALCULATION

  ICRP LUNG MODE

  EXCRETION CURVES (LANGHAM, MOSS, JONES)

COMPLEMENTARY ACTIVITIES

* MEDICAL STUDIES (150 PERSONS BY YEAR)

  BASED ON OCCUPATIONAL PROTOCOLS

  EARLY DISEASE DETECTION OBJECTIVE

  IMPROVEMENT OF SOCIAL ASSISTANCE

  KNOWLEGE OF DEATH CUASES

METEOROLOGICAL STUDIES

OBJECTIVES: TO OBTAIN INFORMATION IN ORDER:

- TO SUPPORT THE SURVEILLANCE PROGRAM IN THE AREA
- TO IDENTIFY LOCAL PHENOMENA (ANNUAL FREQUENCY AND DISTRIBUTION)
- TO CHARACTERIZE THE RESUSPENSION PROCESS

ACTIVITIES:

* DATA COLLECTION FROM THE METEOROLOGICAL TOWERS
* DATA ANALYSES AND INTERPRETATION
* SOFTWARE DEVELOPEMENT TO OBTAIN SPECIFIC PARAMETERS

PRESENTLY A NEW METEOROLOGICAL STATION WITH SEVERAL LEVELS OF MEASUREMENTS IS BEING INSTALLED DEALING WITH RESEARCH ASPECTS OF RESUSPENSION

MEDICAL STUDIES

OBJETIVE: TO KNOW THE STATE OF HEALTH OF THE POPULATION

* BASED ON MEDICAL SURVEILLANCE FOR THE OCCUPATIONAL EXPOSURES WORKERS (SAFETY GUIDE 7.4 FROM THE SPANISH NUCLEAR SAFETY COUNCIL)

* EARLY DETECTION OBJETIVE

* IMPROVEMENT OF SOCIAL ASSISTANCE

* KNOWLEDGE OF DEATH CAUSES

* SAMPLE SIZE: 150 PERSONS EVERY YEAR

* MEDICAL REGISTER OF POPULATION BASED ON INDIVIDUAL FILES

RESEARCH ON DOSIMETRIC ASPECTS

OBJECTIVE: TO IMPROVE THE KNOWLEDGE ON THE METABOLIC BEHAVIOUR OF Pu AND Am IN THE HUMAN BODY FOR INTERNAL DOSIMETRY REQUIREMENTS

ACTIVITIES:

* DUST CHARACTERIZATION FOR BIOKINETICAL STUDIES

  PHYSICO-CHEMICAL AND MINERALOGICAL CHARACTERIZATION

* IN-VITRO LEACHING EXPERIMENTS

  SHORT AND LONG-TERM DISSOLUTION IN LUNG

* DOSES PER UNIT INTAKE FACTORS

* PARTICIPATION IN DOSE EVALUATION INTERCOMPARISON EXERCICES

TOTAL ANALYSES AND MEASUREMENTS FOR THE PERIOD 1966-1991

	SAMPLES	Pu	Am	TOTAL ALFA
AIR	14144	2482	513	
SOIL	5448	4472	2498	3280
VEGETATION	1976	3279	1817	
ANIMALS	104	152	75	
BIOASSAY	2423	2423	891	

WHOLE BODY COUNTER	1.190	(≃ 800 persons)
MEDICAL SURVEY	2093	(≃ 800 persons)

SAMPLING AND ANALYSES ACTIVITIES. PERIOD 1986-1990

	1986	1987	1988	1989	1990	TOTAL	AVERAGE PER YEAR
SOIL							
SAMPLING	187	73	75	100	50	485	97
Pu ANALYSES	70	100	85	59	70	384	77
Am ANALYSES	115	394	30	0	0	539	108
AIR							
SAMPLING	183	156	156	102	144	741	148
Pu ANALYSES	122	102	131	165	446	966	193
Am ANALYSES	26	35	33	88	182	364	73
VEGETATION							
SAMPLING	61	62	71	71	98	363	73
Pu ANALYSES	65	133	0	262	73	533	107
Am ANALYSES	396	211	125	32	0	764	153
ANIMALS							
SAMPLING	6	11	17	8	2	44	9
Pu ANALYSES	14	13	28	5	3	63	13
Am ANALYSES	14	13	28	5	3	63	13
BIOASSAY							
PERSONS	141	149	151	151	149	741	148
Pu ANALYSES	141	149	151	151	149	741	148
Am ANALYSES	141	149	151	151	149	741	148
WHOLE BODY COUNTER	45	68	20	19	24	176	35

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Paper

AIR CONCENTRATIONS OF ^{239}Pu AND ^{240}Pu AND POTENTIAL RADIATION DOSES TO PERSONS LIVING NEAR Pu-CONTAMINATED AREAS IN PALOMARES, SPAIN

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Abstract—On 17 January 1966, an accident during a refueling operation resulted in the destruction of an air force KC-135 tanker and a B-52 bomber carrying four thermonuclear weapons. Two weapons, whose parachutes opened, were found intact. The others experienced non-nuclear explosion with some burning and release of the fissile fuel at impact. Joint efforts by the United States and Spain resulted in remedial action and a long-term program to monitor the effectiveness of the cleanup.

Air concentrations of ^{239}Pu and ^{240}Pu have been continuously monitored since the accident. The average annual air concentration for each location was used to estimate committed dose equivalents for individuals living and working around the air sampling stations. The average annual ^{239}Pu and ^{240}Pu air concentrations during the 15-y period corresponding to 1966–1980 and the potential committed dose equivalents for various tissues due to the inhalation of the ^{239}Pu and ^{240}Pu average annual air concentration during this period are shown and discussed in the report.

1. INTRODUCTION

ON 17 January 1966, there was an aviation accident above the town of Palomares (Cuevas de Almanzora) in southeastern Spain. As a consequence of this accident, four thermonuclear bombs carried by one of the planes fell, and at impact, the nuclear fuel in two of them partially ignited. This gave rise to the formation of an aerosol, which in varying degrees of magnitude, contaminated approximately a 226-hectare area of underbrush, farmland and urban areas.

The contaminated region and the full extent of the areas corresponding to the different levels of resulting α-radiation contamination measured in the period immediately following the accident are shown in Fig. 1. The points of impact of the two thermonuclear devices (which due to the numbering assigned them, correspond to bombs 2 and 3, respectively), are indicated in Fig. 1. The other two bombs were recovered intact, one in the dry river bed near the mouth of the Almanzora River and the other one in the sea.

Where the total soil surface α-contamination values were greater than 1.18 MBq $\times$ m^{-2} (32 μCi $\times$ m^{-2}), the contaminated vegetation and surface layer of soil (approximately 10 cm deep) were collected, separated and disposed of as radioactive waste. The rest of the contaminated area, devoted to agriculture, was irrigated thoroughly, plowed to a depth of approximately 30 cm, and subjected to a light homogenization to reduce the concentration of radionuclides contaminating the soil surface by diluting them in noncontaminated (below the surface) soil. This procedure reduced the α concentration to levels considered sufficiently safe, so there would not be any unacceptable existing Pu-isotope inhalation risks from resuspension to people living in this area or farming the land.

At the time the decontamination operations in the area were finished by removing the wastes and restoring the farmland, a research program was begun.

To maintain constant reference for the various proposed studies on air, vegetation, soil and persons in the Palomares area, the area contaminated during the accident has been subdivided into three zones. These zones correspond respectively to the two which were greatly influenced by contamination arising from each of the thermonuclear bombs that broke up and partially ignited and to the urban zone in which 90% of the population lives. The two zones corresponding to the impact points have been called 2 and 3, in accordance with the numbering of the bombs which fell in them; the zone containing the urban area has been labelled 5. In Fig. 1, the relative location of each one of these zones and its boundaries are shown.

The purposes of our study of air contamination in the area include:

- Measurement of existing ^{239}Pu and ^{240}Pu concentrations in the air of some zones of the area.

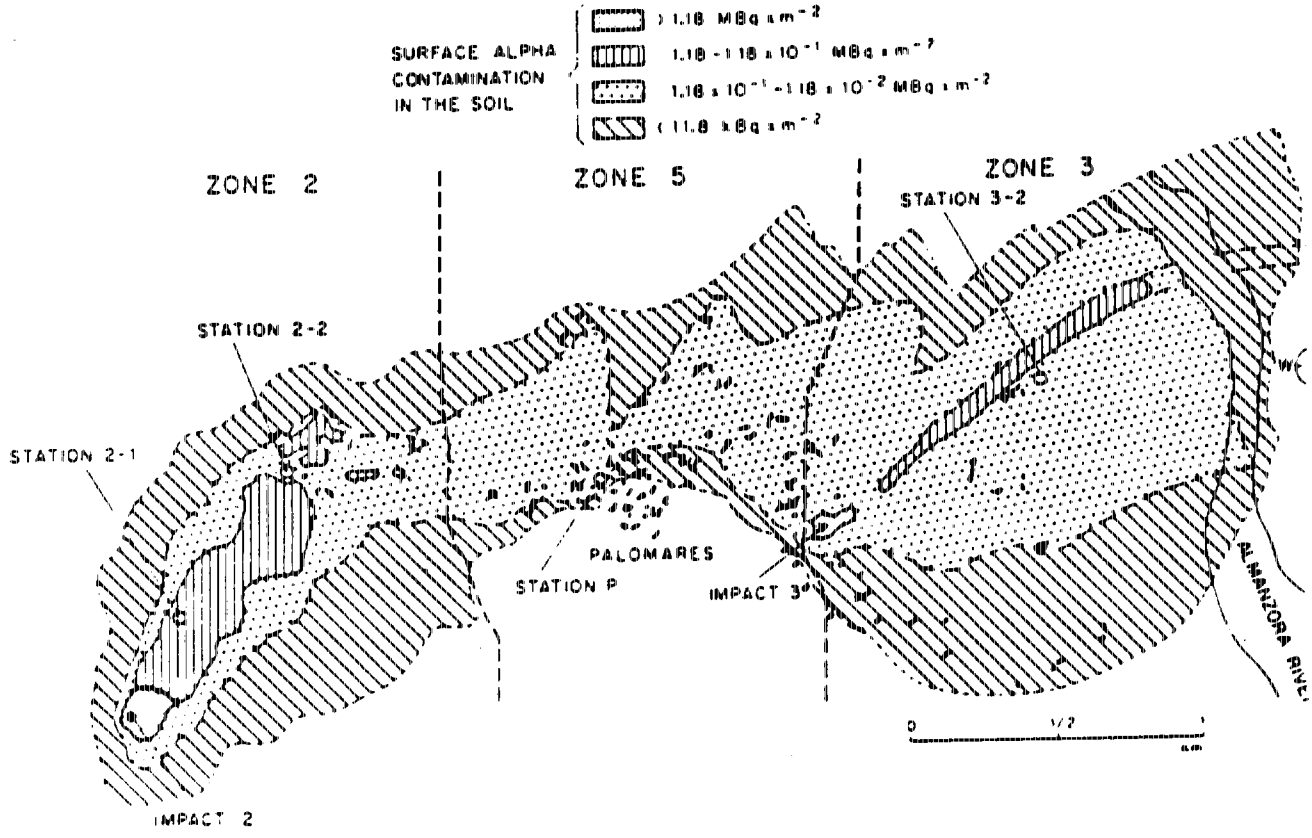

Fig. 1. Palomares area with original surface contamination levels and location of air sampling stations. Black spots show the location of houses.

• Determination of the inhalation risk for people living and farming in the area, and as a consequence, the internal radiation risk for permanent inhabitants, including children.

• Practical deduction about whether the decontamination and restoration actions in the area were correct, and consequently, whether the theoretical reasoning used to estimate the future residual contamination risk was adequate.

In this report the following matters are presented and discussed:

• Average annual ^{239}Pu and ^{240}Pu air concentrations during the 15-y period corresponding to 1966–1980.

• Potential committed dose equivalents for various tissues due to the inhalation of ^{239}Pu and ^{240}Pu average annual air concentration as an aerosol of 1 μm (AMAD) by the ICRP Reference Man during the 15-y period 1966–1980.

2. SAMPLING AND ANALYTICAL PROCEDURES

2.1. *Air sampling*

Throughout the area, four sampling stations were set up—two in zone 2, because it is the most extensive and has greater topographical variation, and one in zc which corresponds to the town, and another in zc In Fig. 1, the location of each is shown. The numbe location of the air sampling stations was selected by t into account the size of the area, its topographical acteristics and the dwelling house distribution.

Characteristics of the area surrounding the four pling stations are as follows:

Station 2-1. The station was located in the sma ley whose boundaries are the hills around the po impact of bomb no. 2. The soil of the nearby surrou area is rocky and has little brush or other wild p Because of these characteristics, plowing was not po The station was located in the center of an area in the surface contamination levels originating from t cident ranged, at that time, from 1.18 MBq × m μCi × m^{-2}) to 1.18 × 10^{-1} MBq × m^{-2} (3.2 μCi ×

Station 2-2. This station is located on a par land devoted to farming and near a small group of dw houses. In this parcel and its environs, surface cor nation levels were between 1.18 × 10^{-1} MBq × m μCi × m^{-2}) and 1.18 × 10^{-2} MBq × m^{-2} (0.32 μCi × However, surface contamination levels up to 1.18 MBq × m^{-2} (3.2 μCi × m^{-2}) occurred in some r areas.

Station P. This station is located in the town and is considered representative of the air breathed by people living in the urban center, as it is in a place that is open to the prevailing winds, which come from the contaminated sectors. The surface contamination in the parcel in which it is located, and those around it, was less than 11.8 kBq × m^{-2} (0.32 μCi × m^{-2}).

Station 3-2. This station was located in the center of a flat strip of cultivated land, which, if the surrounding land near the point of impact is not considered, would have greater contamination due to the impact of bomb no. 3. It is 5 m below the level at which the impact occurred. Surface α contamination of parcels near the sampling station ranged between 1.18 MBq × m^{-2} (32 μCi × m^{-2}) and 11.8 kBq × m^{-2} (0.32 μCi × m^{-2}).

At station 2-2 and P, air samples were taken daily throughout the period 1966–1980; at stations 2-1 and 3-1, samples were collected every day during the period from July 1966 to October 1969. Collecting samples from stations 2-1 and 3-1 were discontinued because of frequent disturbances in the wiring to their power supplies due to farming practices.

Air sampling is continuing and, since 1981, determination of ^{241}Am has been included in the analysis of the samples.

The sampling stations consist of a filter holder, a 1425 rpm, one-third horsepower suction pump, a gas meter for measuring the air volume, and a voltage regulator. Except for the filter holder, this equipment is inside a small wooden booth which protects it from the weather. The filter holder was placed at a height of 1.70 m above the ground.

Filters used are of a cellulose type having a 47-mm diameter and 0.8-μm mesh; the daily aerosol sample taken corresponds to an average air volume of 90 m^3, for the 24-h period.

2.2. *Analyses*

Analyses were performed on composite samples, corresponding to a period of 10 d, for each of the sampling stations. This means that the spectrometric measurement of each compound sample corresponds to an air volume of about 900 m^3.

The radioanalytical technique used to determine the concentration of ^{239}Pu and ^{240}Pu in the aerosol samples basically includes Pu dissolution using nitric acid, separation by ionic exchange, electroplating on stainless steel planchets and α-spectrometric measurement of the α activity.

The analytical technique is as follows:

Filters corresponding to the aerosol samples for each 10-d period are subjected to concentrated nitric acid wet digestion at temperatures of 250°C and 300°C until a white-colored residue is obtained; homogeneous heating is carried out in Al blocks. To calculate the efficiency of the procedures for each sample, 1 mL of standard ^{236}Pu solution, in the early years, and later a ^{242}Pu solution which contains 50 mBq (1.35 pCi) was added.

The residue is dissolved in 50 mL of 8 M nitric acid solution, and then the solution is passed through ion exchange resin* (50–80 mesh) in chloride form, at a flow rate on the order of 1 mL/min. During this phase of the analyses, the Pu and Th are retained in the resin.

To eliminate the small amount of U that the resin might have retained, washing is carried out several times with 8 M nitric acid until a total 150-mL volume is obtained. To eliminate the Th retained in the resin, the column is washed once with 12 N HCl and the wash effluent is discarded.

Some grains of hydroxylammonium-chloride are added to the resin column and the Pu fixed in the resin is eluted by means of three washes with 5 mL of 0.5 N HCl. The effluents are dried at a temperature not to exceed 80°C, dissolved again in the concentrated HCl and are dried again at the same temperature as before. These dried residues are dissolved in 1 N hydrochloric acid.

Four percent ammonium oxalate is added to the 1 N hydrochloric acid solution and it is subjected to electrolysis. The Pu is electroplated onto the 14-mm-diameter stainless steel planchet at a power of 20 V and a current intensity of 200 mA for 3 h.

The planchets are measured by α spectrometry with Si barrier detectors having an active area of 300 mm^2 and a multichannel pulse height analyzer. The counting efficiency of the equipment used is on the order of 31%.

The detection limit of the method is 1.8 μBq × m^{-3} (5 × 10^{-2} fCi × m^{-3}), which is equivalent to 10^{-5} of the derived air concentration (DAC) for ^{239}Pu and ^{240}Pu compounds of Class Y to radiation workers.

3. RESULTS AND DISCUSSIONS

The results of ^{239}Pu and ^{240}Pu concentrations in samples taken at the air sampling stations and the potential committed dose equivalents to various tissues in the scenario of continuous exposure to inhalation of the average annual concentrations by the dwellers of Palomares are discussed below.

3.1. *Air concentration of* ^{239}Pu *and* ^{240}Pu

In accordance with the annual limit on intake (ALI), the DACs for ^{239}Pu and ^{240}Pu recommended by the International Commission on Radiological Protection (ICRP) in its *Publication No.* 30, for radiation workers exposed 2000 h per working year are the following:

Class Y compounds: 200 mBq × m^{-3} (5.4 pCi × m^{-3})

Class W compounds: 80 mBq × m^{-3} (2.16 pCi × m^{-3}).

As the ICRP still has not established ALI and DAC values for the public and has suggested the use of a fiftieth or a hundredth of the values for established radiation workers, we have decided to use values that could be more

* Dowex AG-1X-2, Bio Rad Laboratories, 32nd & Griffin, Richmond, CA.

or less equivalent to those that might be recommended in the future by the ICRP.

People in the Palomares area should be considered individually among the continuously exposed public, that is, 8,760 h y^{-1}.

Taking into consideration the ratio between annual dose limits recommended for workers and members of the public by the ICRP, 10:1, and the continuous exposure of the public in the area (an adult Reference Man breathing volume = 23 $m^3 \times d^{-1}$), it has been calculated that the DACs of ^{239}Pu and ^{240}Pu for members of the public are the following:

Class Y: 5.9 mBq × m^{-3} (0.16 pCi × m^{-3})

Class W: 2.4 mBq × m^{-3} (0.06 pCi × m^{-3}).

Because we are dealing with people living in an urban zone and some of them will continue to live there for their lifetime in agricultural work, we applied the ICRP recommendation that, in such cases, the annual effective dose equivalent limit should be reduced so that the effective dose equivalent throughout their lives corresponds, at a maximum, to the value resulting from receiving a 1-mSv (0.1 rem) mean annual effective dose equivalent rate. Consequently, we adopted the following values for the DAC, which we designate DAC (general public) in this report.

Class Y: 1.2 mBq × m^{-3} (32 fCi × m^{-3})

Class W: 0.5 mBq × m^{-3} (13 fCi × m^{-3})

We believe that the Pu aerosols are found as ox PuO_2, such as were formed during the burning of the monuclear bombs at impact. Consequently, the cor sions, insofar as real existing risk is concerned, wil those derived from the concentration of ^{239}Pu and and the DAC for Class Y compounds.

A comparative study to consider the presence of C W compounds was conducted to assure ourselves maximum protection was being provided.

The analyses of the values obtained for the ^{239}Pu ^{240}Pu concentrations in the air samples taken in the ornares area during the period 1966–1980 and their c parison with the DAC limits set forth in the prece paragraphs allow us to make the following observat and conclusions:

(a) In the air samples taken at the four samp stations, the frequency of ^{239}Pu and ^{240}Pu particles in centrations exceeding the detection limit, 1.8 μBq × (0.05 fCi × m^{-3}), are shown in Fig. 2. From the va shown in Table 1, it can be concluded that:

- Plutonium-239 and ^{240}Pu particles are resuspen in the air. The presence of such particles has been es lished in samples taken at the four stations.
- The frequency of the existence of Pu has b varying as a function of time for each one of the samp stations.
- The sampling station location with regard to sidual contamination, taken as the average annual centage of concentrations greater than the detection li during the period 1966–1969, was 37.0 in the urban z

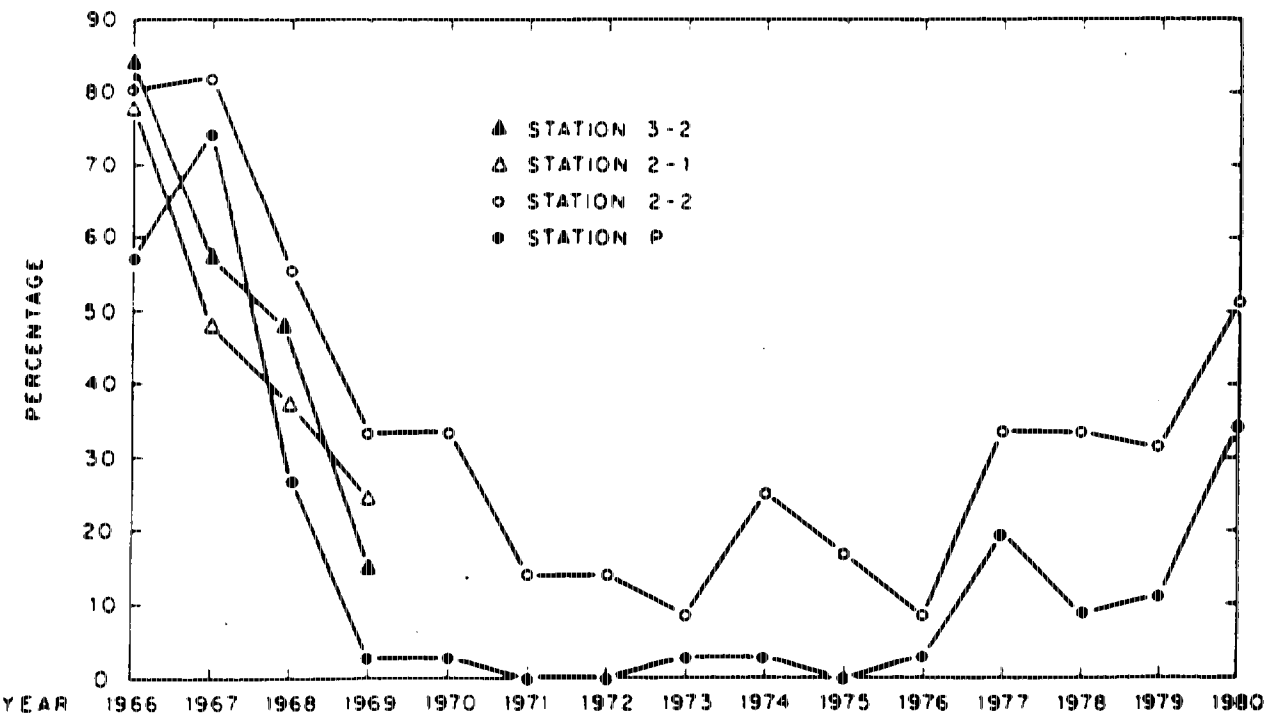

Fig. 2. Percentage of air samples with a ^{239}Pu and ^{240}Pu concentration exceeding the detection limit.

Table 1. Percentage of air samples with a ^{239}Pu and ^{240}Pu concentration exceeding the detection limit (1.8 uBq × m^{-3}) during the period 1966–1980

YEAR	AIR SAMPLING STATION			
	2-1	2-2	P	3-2
1966	77.8	80.0	57.1	84.2
1967	47.8	81.8	74.3	57.1
1968	37.1	55.6	26.5	48.0
1969	24.0	33.3	2.8	14.8
1970		33.3	2.8	
1971		13.9	0	
1972		13.9	0	
1973		8.3	2.8	
1974		25.0	2.8	
1975		17.1	0	
1976		8.3	2.8	
1977		33.3	19.4	
1978		33.3	8.8	
1979		31.4	11.1	
1980		51.4	34.3	
TOTAL	42.7	33.0	14.7	49.1
TOTAL 1966–1969	42.7	60.0	37.0	49.1

(station P), while at station 2-2, it was 60.0; values corresponding to these average annual percentages at stations 2-1 and 3-2 were 42.7 and 49.1, respectively.

• For samples taken at station 2-2 and station P during the period 1966–1980, the average annual percentage of concentrations greater than the detection limit was 33 at station 2-2, while at station P it was 14.7.

At station 2-2 and station P, the maximum values for the sampling percentage with a ^{239}Pu and ^{240}Pu content in excess of the detection limit occurred in 1967, with values of 81.8 and 74.3, respectively. At station 2-2, minimum percentages of samples having a ^{239}Pu and ^{240}Pu content in excess of the detection limit occurred in 1973 and 1976, with a value of 8.3 in both cases. As of 1977, an increase in values began and reached a value of 51.4% by 1980. This means that as of 1977, the resuspension of existing ^{239}Pu and ^{240}Pu in the soil of zone 2 increased, as evidenced by the fraction of measurements above the detection limit at station 2-2. The resuspension increase was, as can be proven, related to earth moving without previous watering on farmlands located in parts of zone 2, which either had not been cultivated since the accident or never had been cultivated, because it is hilly terrain covered with poor-quality, wild vegetation.

As of 1968, the number of samples at station P with a ^{239}Pu and ^{240}Pu content greater than the detection limit diminished considerably, so that between 1969 and 1976, percentages of samples with a content exceeding the detection limit were between 2.8 and 0. As of 1977, and in the case of station 2-2, an increase in these percentages occurred, reaching a value of 34.3 in 1980, which we think was due to earth movement, especially in zones 2 and 5, in order to convert uncultivated land into cultivated land and to build greenhouses.

(b) Average annual concentrations of ^{239}Pu and ^{240}Pu have been assessed via the 10-d composite samples taken every year at each sampling station. The values for the average annual ^{239}Pu and ^{240}Pu air concentration during the 15-y period 1966–1980 for stations 2-2 and P are those shown in Table 2. Average annual concentrations for stations 2-1 and 3-2 during the period 1966–69 are also shown.

To be conservative, we assumed that all Pu is respirable with an AMAD of 1 μm and no account has been made for the Pu in the sample arising from nuclear fallout.

From the air concentration values shown in Table 2, we can arrive at the following:

• Most of the average annual concentrations of ^{239}Pu and ^{240}Pu in the air have been less than a tenth of the calculated DAC (general public) of Class Y Pu compounds [1.2 mBq × m^{-3} (32 fCi × m^{-3})], at all the sampling stations and in all years studied. The exceptions correspond to air sampling station 2-2 (1967 and 1969) and air sampling station 2-1 (1969).

• With the exception of 1 y (1967) at station 2-2, the average concentration values have also been less than the DAC (general public) values for Class W Pu compounds [0.5 mBq × m^{-3} (13 fCi × m^{-3})].

• At station P, whose values are representative of the exposure of the Palomares dwellers, the maximum

Table 2. Average annual concentrations of ^{239}Pu and ^{240}Pu in the air during the period 1966–1980

YEAR	CONCENTRATION AT THE STATION (μ Bq × m^{-3})			
	2-1	2-2	P	3-2
1966	41.8	44.8	14.8	27.4
1967	15.2	441.8	4.1	13.0
1968	7.0	21.8	2.6	3.3
1969	161.0	142.1	2.6	14.1
1970		5.9	2.2	
1971		2.2	<1.8	
1972		10.4	<1.8	
1973		3.0	2.2	
1974		8.1	4.1	
1975		16.3	<1.8	
1976		4.4	<1.8	
1977		11.8	5.6	
1978		16.7	2.2	
1979		19.2	5.6	
1980		32.9	2.8	

average annual concentration was in 1966, 14.8 μBq $\times$ m^{-3} (0.4 fCi $\times$ m^{-3}), and then a decline occurred, which for the period 1968 to 1976 remained more or less constant.

As of 1977, the annual concentration of activity increased, and in 1980 it reached the maximum values for the entire period, 28 μBq $\times$ m^{-3} (0.76 fCi $\times$ m^{-3}). This increase was, as previously indicated, related to earth movement without previous watering to reshape a part of the hilly terrain with residual surface contamination corresponding to zone 2 and some tracts of uncultivated land corresponding to zone 5 into cultivable parcels.

Values from station 2-2 are representative of the double influence of working contaminated land (resuspension) and of some persistence of some of the original contamination in the hilly zone, because the slope and large number of rocks prevented plowing during the original remedial action activities. The maximum average annual concentration occurred in 1967, with a value of 442 μBq $\times$ m^{-3} (11.9 fCi $\times$ m^{-3}), coinciding with renewed cultivation of the zone after the accident. Between 1970 and 1976 stabilization of the activity concentration occurred with relatively small fluctuation. Beginning in 1977, and as in the case of station P, the annual concentration of activity increased again, reaching a value of 33 μBq $\times$ m^{-3} (0.89 fCi $\times$ m^{-3}) in the year 1980.

Values for the ^{239}Pu and ^{240}Pu air concentration corresponding to the entire period, and especially the 4 y during the period 1977–1980, were generally greater at station 2-2 than at the others. These were related to the residual contamination in the hilly area and to proximity to the area in which earth movement occurred as of 1977.

The maximum value for a single 10-d ^{239}Pu and ^{240}Pu composite air concentration corresponds to station 2-2, 9.3 mBq $\times$ m^{-3} (0.25 pCi $\times$ m^{-3}); this value occurred during the month of August 1967, and was the only one exceeding the DAC for the public of Class Y Pu compounds [5.9 mBq $\times$ m^{-3} (0.16 pCi $\times$ m^{-3})].

Concentrations exceeding the DAC (general public) of Class Y Pu compounds [1.2 mBq $\times$ m^{-3} (32 fCi $\times$ m^{-3})] only have been produced during 1966–69 in four 10-d periods, three of which corresponded to station 2-2 and one to station 2-1. The latter occurred in 1969, and the others corresponding to station 2-2 in the years 1967 (two 10-d periods) and 1969 (one 10-d period). It should be recalled that the DAC (general public), as used in this paper is about 170 times below that used by ICRP for workers.

Concentrations exceeding the DAC (general public) of Class W Pu compounds occurred during the period 1966–69 in five 10-d periods. Of these, one corresponded to station 2-1, and four to station 2-2. During the period 1970–1980 only on one occasion, at station 2-2, was this DAC (general public) exceeded.

• The mean values for average annual concentrations of ^{239}Pu and ^{240}Pu in air for each of the sampling stations during the period 1966–69 were the following:

Station 2-1: 56 μBq $\times$ m^{-3} (1.5 fCi $\times$ m

Station 2-2: 160 μBq $\times$ m^{-3} (4.4 fCi $\times$ r

Station P: 6 μBq $\times$ m^{-3} (0.18 fCi $\times$ m

Station 3-2: 14 μBq $\times$ m^{-3} (0.39 fCi $\times$ r

The average air concentration values of ^{240}Pu during the entire period 1966–1980 we lowing:

Station 2-2: 52 μBq (1.4 fCi $\times$ m^{-3})

Station P: 5.5 μBq (0.15 fCi $\times$ m^{-3}

By comparing the average concentration ing each of the periods studied at each of the stations with the DAC values, one can conclu

• In the urban area, P, the average conc both for the period 1966–69 as well as the per 1980, has been below a hundredth of the DA public) for Class Y Pu compounds and belo entieth of the DAC (general public) for Class W pounds.

• In the farming zone, 2-2, the average con for the period 1966–1980 was below a tenth o (general public) for Class Y Pu compounds a order of a tenth of the corresponding DAC (gene for Class W Pu compounds. During the perioc the average concentration also was below the I eral public) for Class Y and Class W compour

• During the period 1966–69, the avera tration in sampling stations 2-1 and 3-2 was bel of the DAC (general public) of Class Y Pu c and below the DAC (general public) of Class W pounds.

3.2. *Potential committed dose equivalents to*

For sampling stations P and 2-2, which sentative of the risk in the urban center anc significant farming area (critical group zone), w culated the committed dose equivalents rece assumed inhalation of the average annual ^{239}Pu concentrations shown in Table 2 for the per 1980. The committed dose equivalents to vari and tissues have been calculated by means of Sv/Bq, for committed dose equivalents per u of inhalation intake recommended by the In Commission on Radiological Protection (ICR K. F. Eckerman (Ec81).

The potential committed dose equivalen and a mean aerosol size (AMAD) of 1 μm ar Figs. 3 and 4 for each year. In Fig. 3 the comm equivalents to lungs, bone surface, liver, red bor and gonads of people in the urban zone are gi 4 the values for people in the cultivated zon greatest potential risk, station 2-2, are shown.

From the discussion of values calculated fc gans the following has been deduced:

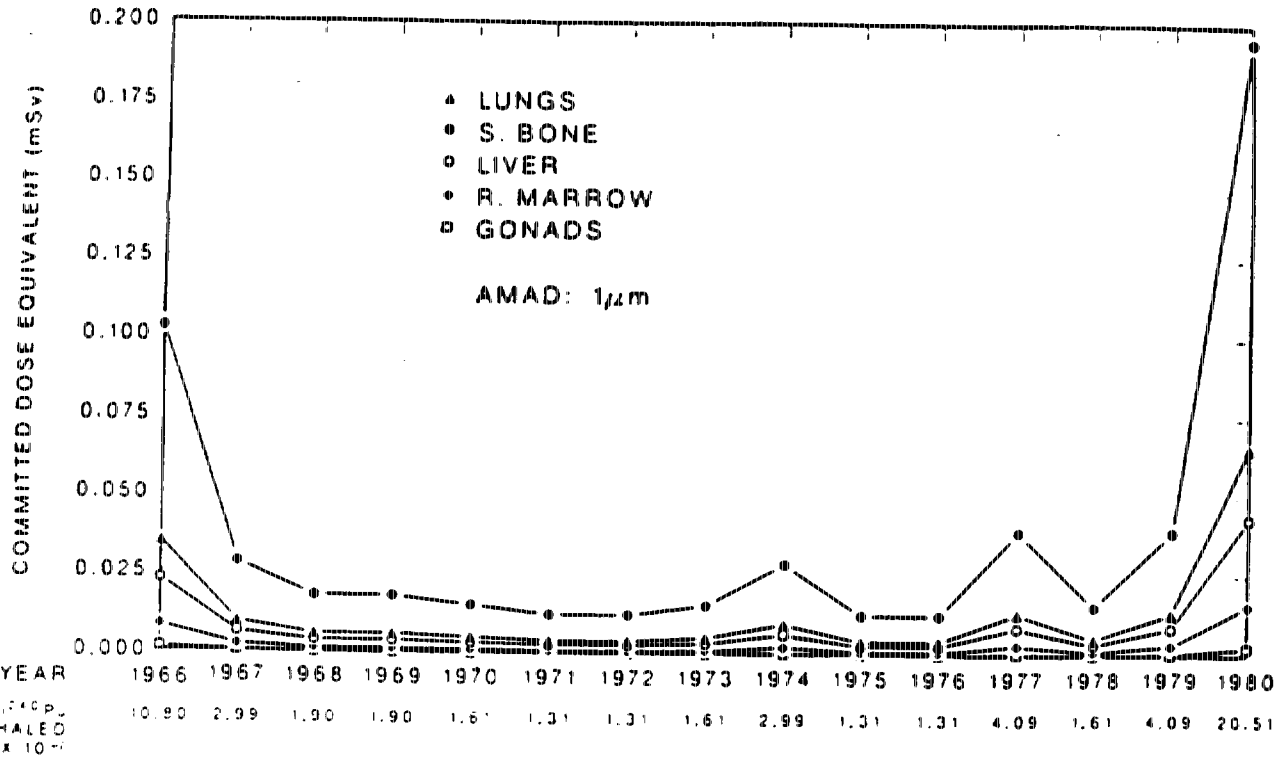

Fig. 3 Potential committed dose equivalents for adults in urban area from inhaling the average annual ^{239}Pu and ^{240}Pu concentration during the period 1966–1980.

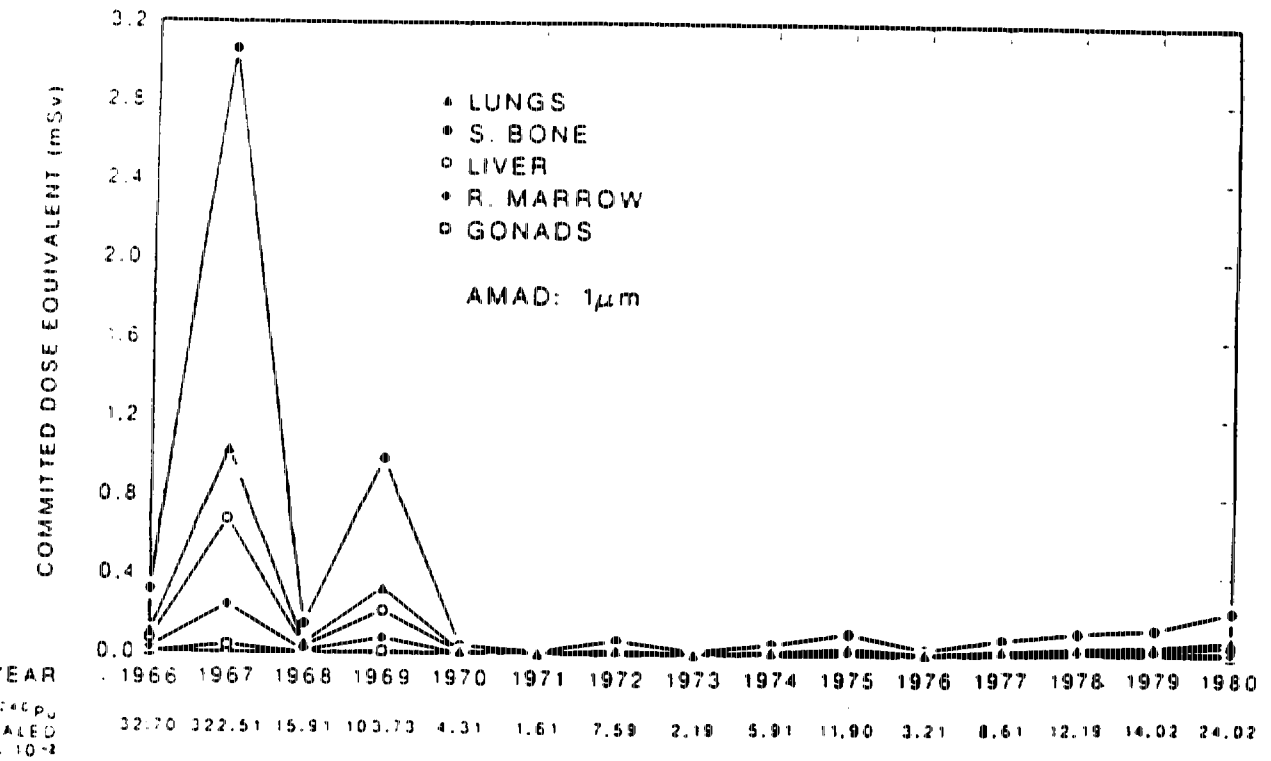

Fig. 4. Potential committed dose equivalents for adults in the "Critical Group" area from inhaling the average annual ^{239}Pu and ^{240}Pu concentration during the period 1966–1980.

• The potential committed dose equivalent to bone surfaces is the highest and the summation of the committed dose equivalents for each of the 15 y during the period 1966–1980 shows a value of 0.5637 mSv (56.37 mrem) for people in the urban zone and 5.4187 mSv (541.9 mrem) for people in the zone around station 2-2. Its contribution to the committed effective dose equivalent to people during the 15 y will be 0.0169 mSv (1.69 mrem) and 0.1625 mSv (16.25 mrem) at each zone, respectively.

The total potential committed dose equivalents for the other organs and their contribution to the committed effective dose equivalent during the period 1966–1980, at each zone, are shown in Tables 3 and 4 respectively.

• The ICRP has recommended a value of 1 mSv (0.1 rem) for the effective annual dose equivalent limit for individuals in the population who might be exposed over a long period of their lives. Bearing this in mind, the annual limiting dose equivalent value for each particular organ, considering its exposure to the exclusion of the others and keeping in mind the organ weighting factor for stochastic effects, is 33.33 mSv (3333 mrem) for the bone surface, 8.33 mSv (833 mrem) for the lungs, 16.67 mSv (1667 mrem) for the liver, 8.33 mSv (833 mrem) for red bone marrow and 4 mSv (400 mrem) for gonads.

On comparing the values of the committed dose equivalents calculated for each organ with those deduced and presented in the previous paragraph for the separate exposure of each organ, and in considering the exclusion of the others, we can observe that the total committed dose equivalent values for each organ during the 15-y period are less than the annual limits for the organ.

• The contribution of the committed dose equivalents in the five organs to the potential committed effective dose equivalent to people during the 15-y period 1966–1980 is 0.0544 mSv (5.44 mrem) in the urban area and 0.5224 mSv (52.24 mrem) in the area which corresponds to the critical group.

4. CONCLUSIONS

From the discussion of the results given in the previous sections, the following conclusions can be deduced:

(1) With the exception of the urban zone during 1971, 1972, and 1975, at all sampling stations and for all the years studied, ^{239}Pu and ^{240}Pu concentrations in the air have been found on occasion to exceed the detection limit, 1.8 μBq × m^{-3} (5 × 10^{-2} fCi × m^{-3}). No corrections were made for the contribution to the measurements from worldwide nuclear weapons fallout.

Table 3. Total potential committed dose equivalents for organs during the period 1966–1980

ORGAN	TOTAL COMMITTED DOSE EQUIVALENT, mSv	
	URBAN ZONE (P)	STATION 2-2
BONE SURFACE	05637	54187
LUNGS	01902	18254
LIVER	01250	11978
RED BONE MARROW	00450	04336
GONADS	00071	00684

Table 4. Contribution of committed dose equivalents in organ to the total potential committed effective dose equivalent during the period 1966–1980

ORGAN	CONTRIBUTION TO COMMITTED EFFECTIVE DOSE, mSv	
	URBAN ZONE (P)	STATION 2-2
BONE SURFACE	00169	01625
LUNGS	00228	02190
LIVER	00075	00716
RED BONE MARROW	00054	00520
GONADS	00018	00171

The frequency of air samples containing ^{239}Pu and ^{240}Pu concentrations greater than the detection limit progressively diminished with time following the accident. During the period 1970–76, the frequency of measurements above the detection limit stabilized except for station 2-2 in 1974. During this period, agricultural operations were carried out normally throughout the zone, except for a hilly and uncultivated parcel near station 2-2; it was plowed and transformed into a cultivated parcel during 1974.

During 1977–1980, there was an increase at stations 2-2 and P in the frequency of samples with ^{239}Pu and ^{240}Pu concentrations in the air greater than the detection limit, especially at station 2-2. This increase was related to the major earth moving activities (leveling and terracing) of land that never had been cultivated (or at least, never after the accident) into cultivable land. This land corresponded to the hilly area in zone 2. Analogous changes were carried out during 1979 and 1980 on tracts of uncultivated land located in the zone near the urban center.

(2) The average annual ^{239}Pu and ^{240}Pu concentration in the air during the sampling periods corresponding to each of the stations has been below the DAC (general public) for Class Y Pu compounds [1.2 mBq × m^{-3} (32 fCi × m^{-3})]. Most of them have been below one tenth of the DAC (general public); the exceptions correspond to air sampling station 2-2 (1967 and 1969) and air sampling station 2-1 (1969).

(3) The average ^{239}Pu and ^{240}Pu concentration in air to which the public living in the urban zone has been exposed during the 15-y period (1966–1980) is less by a factor of 4.6 × 10^{-3} of the DAC (general public) of Class Y Pu compounds and by a factor of 1.1 × 10^{-2} for those of Class W.

In this urban zone the average annual ^{239}Pu and ^{240}Pu concentrations always were below those corresponding to the other sampling stations for the same year; maximum concentration was in 1980, with a value of 28 μBq × m^{-3} (0.76 fCi × m^{-3}).

(4) At sampling station 2-2, located between cultivated parcels near the hilly area and which gave evidence of higher residual surface contamination, the highest average annual concentrations have occurred throughout the period, with the exception of 1969. The maximum concentration occurred in 1967, with a value of 442 μBq $\times$ m^{-3} (11.9 fCi $\times$ m^{-3}). Thus, most values of these average annual concentrations always were below a tenth of the DAC for the public of Class Y Pu compounds.

At this sampling station, the maximum Pu concentration in the air for the entire period was detected, and it had a value of 9.3 mBq $\times$ m^{-3} (0.25 pCi $\times$ m^{-3}), which corresponded to a 10-d sampling period during the month of August 1969.

(5) The contribution of the potential committed dose equivalents in the organs considered (lungs, bone surface, liver, red marrow and gonads) to the potentia committed effective dose equivalent received by peopl in the area by continuous inhalation during the perio 1966–1980 is 5.44 $\times$ 10^{-2} mSv in the urban zone an 5.23 $\times$ 10^{-1} mSv (52.3 mrem) in the zone correspondin to the "critical group." These values correspond to 5.44 and 52.34% of the effective dose equivalent limit of 1 mS (0.1 rem) per year, that is 0.36% and 3.48% of the tota effective dose equivalent limit for a 15-y period.

Acknowledgments—We wish to express our appreciation to the U.S Department of Energy and its predecessor agencies for help in carryin out this work, as well as to the technicians of the Junta de Energia Nuclear José Maria Montero, Mariano Casado, Camila Blanco and Francisc Moreno. We also wish to thank C. R. Richmond, Oak Ridge Nationa Laboratory, Oak Ridge, TN, for advice and assistance.

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EVALUATION OF REMEDIAL ACTIONS TAKEN IN AGRICULTURAL AREA CONTAMINATED BY TRANSURANIDES

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1. INTRODUCTION

In January 17th, 1966 while flying over Palomares (Almería) in the Southeast of Spain two United States Air Force planes collided while on a mid-air fuel supply operation. One of the planes carrying four thermonuclear weapons, three of which, one intact were found on land in the vecinity of Palomares, 24 hours after the destruction of the planes. The fourth thermonuclear weapon was found in the Mediterranean Sea in April 7th. The parachutes of two bombs did not function, causing the detonation of the conventional explosives within them. Thus, releasing their fisionable material and igniting part of it. The aerosol formed by the ignition of the fisionable material produced a contaminating cloud which covered 226 hectares (558 acres) of uncultivated farm and urban land. (See figure 1).

Bomb number 2, for the purpose of this study, landed about one mile to the West of Palomares (impact point number 2) and the plutonium - bearing dust cloud was carried by 30-knot West winds, over non-cultivated terrain, irrigated fields and the northern edge of the village. The cloud from impact point 3,located in the east edge of the village, traveled away from the village but across farmed areas.

An assessment of the situation began shortly after the crash. Visible fragments of both, bombs and air crafts were recuperated, proceeding to determine the levels of alpha contamination in soil, vegetation, houses and area residents. Superficial alpha contamination levels were measured with PAC-15 alpha detectors.

Figure 1 shows the various levels of surface alpha contamination produced and the extension corresponding to each level of alpha contamination. The highest levels of contamination were found in non-cultivated lands located between small hills 1.500 meters southeast of town.

Contaminated vegetation was recuperated, treated and considered as radioactive waste.

Table 1 shows remedial actions taken regarding the affected lands according to the knowledge of the time so there would not be any unacceptable risk in the short and long term contamination of the crops in the area. Ten centimeters of top soil were removed in the areas were superficial alpha concentration was 1200 kBq/m^2 or higher. This material was sealed in containers and sent to the United States. Arable land with levels below 1200 kBq/m^2 were wet down, plowed to a 30 cm depth, harrowed and mixed. On rocky hillsides in area 2, where plowing was not possible, soil with plutonium level above 120 kBq/m^2 was removed in some degree by hand tools.

TABLE 1. REMEDIAL ACTIONS TAKEN AT PALOMARES

LOCATION	SURFACE	REMEDIATION
Impact point 2	1.6 Ha	Removed top 10 cm
Impact point 3	0.6 Ha	Removed top 10 cm
Remainder	224 Ha	Plowed to 30 cm where possible

Once remedial actions were completed and radioactive wastes were removed, an experimental radiological surveillance program was established in order to study long term environmental effects.

Figure 1 shows the location of the six 50 x 50 m study plots established in order to comply with the objectives of the short and long term investigation program on residual plutonium contamination on lands and their effect over farming crops produced in the areas. These study plots have been yearly surveyed since cultivation began to determine contamination by $^{239}Pu+^{240}Pu$ of agricultural products. However, since the study plots have not been cultivated yearly due to diverse reasons, since 1978, sampling has been extended to neighboring areas where residual $^{239}Pu+^{240}Pu$ was similar.

Farming procedures in the area are typical of Mediterranean agricultura zones of scarce yearly precipitation in the order of 200 l/m^2 which require artificial means of irrigation. Until recent years irrigation by flooding with water pumped from wells in the area was prevailing system. Recently, specially, in the production of tomatoes, melons, etc. a drop to drop system is used. For cereals and alfalfa flooding procedures and sometimes aspersion systems are still in use.

This report deals specifically with the evaluation of the remedial actions taken in the area, through the incidence of plutonium contamination in agriculture products produced in the area, and, therefore, the risks to individuals derived from the use of this products through their direct and indirect consumption, ie. animal feed.

Towards this end, we shall expose the results of the field work and their application in order to determine the dose, which has allowed us to arrive to some conclusions in relation to:

a) Evolution of $^{239}Pu+^{240}Pu$ concentrations in soil of cultivated plots.

b) Concentration of $^{239}Pu+^{240}Pu$ in the areas main crops (tomatoes, barley and alfalfa) through a long period of time.

c) Relation of $^{239}Pu+^{240}Pu$ concentrations in soils and vegetation in order to estimate transference factors due to agricultural practice typical in the area and its weather conditions.

d) Concentration of $^{239}Pu+^{240}Pu$ in the totality of annual crops by hectare for each principal crop, and in consideration of the estimated transference ratios.

e) Collective committed effective dose equivalent, $S_{E,70}$, resultant from direct ingestion by humans of annual tomato crops by a cultivated hectare, and in meat, milk of cows, pigs, etc. consuming animal feed from the area.

f) Collective risk derived from the remedial actions taken in the area and deduction of intervention levels towards the application of these remedial actions in the agricultural zones and with similar crops and characteristics as in Palomares, affected by plutonium contaminated aerosols.

2. FIELD EXPERIMENTS AND RESULTS

The field experimentation carried out along the years and the results obtained are expounded below.

2.1 PLUTONIUM CONCENTRATION IN SOILS.

The soils of Palomares, according to their petrographical composition can be classified as lithograywacke-phylloarenite with a 50% matrix where the granular component (grave-sands) are fragments of rocks, mainly shale-type metamorphic rocks.

The average mineralogical composition is: 38.5% quarz, 39.0% moscovite-illite, 21.4% carbonates (calcite and dolomite), 3% iron oxides and opaque minerals, 1.4% chlorite and 2% vegetable remains. These components represent more than 95% of the total mineral composition.

The medium content of organic carbon is 0.27% and humic acids represent 47% of the total organic carbons.

In order to study the evolution of Plutonium concentration in the soil due to the agricultural activities in the areas on a long term basis, six study plots (figure 1) were established. These plots were designated as 2-1, 2-2, 5-1, 5-2, 3-1, 3-2. In order to determine the concentration of plutonium in these plots, periodic sampling has been conducted since 1966 from nine points along the diagonals, equidistant from each other. Each soil sample was 30 milimeter diameter and 45 centimeter deep divided into five sections (0-5, 5-15, 25-35 and 35-45).

Plutonium concentrations in soil samples represent a total of 2160 analysis. The results have shown that plutonium concentrations are heterogenous, but due to farming practices over the years, homogeneity

is increasing, always showing a slightly variable concentration in the top soil.

Table 2 shows the average concentration of $^{239}Pu+^{240}Pu$ in each of the study plots. Due to the fact that plot 2-1 has not been cultivated the plutonium concentration shown corresponds to the top 5 cm.

In all study plots a correlation between the size of soil particles and the concentration of plutonium exist. The largest concentration corresponds to the fraction between 63 and 250 μm. Fractions smaller than 10 μm have only been associated with 15% of the total plutonium activity, increasing the associated percentage in particles of less than 5 μm in the more cultivated terrains.

2.2 PLUTONIUM CONCENTRATIONS IN CULTIVATED CROPS.

The main crops in the area are tomatoes, barley and alfalfa. In the past few years the production of water melons and peppers increased. Corn, beans and other products are only a small percentage of the areas cropped. Thus, our main interest in this report has been the correlation between plutonium concentrations in soil and contamination of tomato, barley and alfalfa crops in the area.

The plan to determine plutonium contamination in the area began in 1968. It sampled and analyzed agricultural products cultivated in the plots used to study the evolution of residual plutonium in the soil.

Vegetation sampling took place within an area of a circunference of a one meter radius, with a center in each of the nine points in the plot where the soil samples were taken. Each sample correspond to a 5-10 kg of weight. When control of other plots in the zone was established in 1978, a radon sampling system was implemented taking 5-10 kg samples of each part of the studied plants.

Many years of sampling have demostrated that not all samples studied, even from the same plot, present plutonium contamination higher than the minimum concentrations detected by our analytical procedures.

Table 3 shows the number of samples analyzed, the percentage of those which show higher than minimum detectable concentration of plutonium and $^{239}Pu+^{240}Pu$ concentration in each of the parts of the agricultural products controlled; this average concentration corresponds to the results of higher minimum detectable concentrations.

As a result of the observation of the samples labeled positive we can deduce that the effect of resuspension of the plutonium particles in soil plays an important role in the contamination of agricultural products cultivated in the area. The highest percentage corresponds to those parts of the plants which present larger surface or higher posibilities of retaining superficial particles (tomato leaves, straw and barley spicules, alfalfa leaves). We shall also considerer important that only 6% of the samples of washed tomatoes show plutonium contamination. Thus, we considerer that a great part of the plutonium contamination in these agricultural products is due to the external surface contamination and not by absorption through the plants'root.

Tables 4, 5, 6 and 7 show the average concentration of $^{239}Pu+^{240}Pu$ in tomatoes, barley and alfalfa, corresponding to the samples taken through long periods of time in each of the estudy plots and neighboring areas. These tables also express the values of soil plant concentration ratios. According to these values, and for each and every crop, we can deduce the following:

a) Tomatoes.

The average concentration of $^{239}Pu+^{240}Pu$ in the edible part are comprised between 0.04 and 0.45 Bq x kg^{-1} and are inferior in one order of magnitude to those of the plants, stalk and leaves, which have shown concentration of $^{239}Pu+^{240}Pu$ in the range of 0.04 to 7.68 Bq x kg^{-1}. In washed tomatoes, we have found lower percentage of contaminated samples on top of which the concentration of $^{239}Pu+^{240}Pu$ is in a range of 0.004 to 0.24 Bq x kg^{-1}.

The soil-fruit concentration ratio are in the range 0.38×10^{-4} - 2.70×10^{-4}. The soil-plant concentration ratio are between 0.04×10^{-3} and 5.24×10^{-3}.

b) Barley.

Contamination by $^{239}Pu+^{240}Pu$ are different in the diverse parts of the barley plant, which are not generally higher in a scale of magnitude. The average concentration of $^{239}Pu+^{240}Pu$ in the barley grain ranges between 0.34 and 6.00 Bq x kg^{-1}. In the straw it ranges between 1.36 and 12.51 Bq x kg^{-1} and in the spicule between 0.65 and 10.35 Bq x kg^{-1}.

The soil-grain concentration ratios determine range between 2.9×10^{-4} to 4.34×10^{-3}. For the straw, the soil-straw concentration ratio ranges between 2.14×10^{-3} to 1.32×10^{-2}. For the spicule, which is the part of the barley plant not used in animal feed since it is discarded during the preparation process, the soil-spicule concentration ratios are in the range of 5.9×10^{-4} to 1.97×10^{-2}.

c) Alfalfa.

The average concentration of $^{239}Pu+^{240}Pu$ corresponding to samples of edible parts of the alfalfa obtained in the diverse plots of the area range between 0.90 and 40.30 Bq x kg^{-1}. The values for soil plants concentration ratios range, except for one sample set, between 1.78×10^{-3} to 8.2×10^{-2}.

The range of values corresponding to soil plant concentration ratios obtained from the samples with concentrations of $^{239}Pu+^{240}Pu$ higher than our detectible minimums, and the percentage of samples with positive results, included in Table 3, show to our judgement that the resuspension plays an important role in plutonium contamination in crops produced in the area of which a significant part is of the external surface type.

As a result of the experimental data collected in the field work carried out in the Palomares area, we have concluded that given its climatological conditions and the farming procedures used for each type of crop, the average values of soil-plants concentration ratios are those expressed in Table 7.

2.3 DOSES DERIVED FROM THE CROPS.

Based on the medium of the data obtained yearly regarding the productivity of the crops in the Palomares area, it can be deduced that the annual harvest is the following:

Tomatoes: 80.000 Kg/Ha

Barley:

Grain: 2.500 Kg/Ha

Straw: 2.200 Kg/Ha

Alfalfa: 70.000 Kg/Ha

We have considered to take the maximum conservative stand, which points to the fact that all harvest is contaminated, even though it has been stated that this is not so.

Table 8 shows the values of the collective committed effective dose equivalent. These have been determine after considering the average soil-crop concentration ratios (Table 7), the ingestion to milk transfer coefficients specified for Terra Code (ref. 1) and the highest values of Sv/Bq ratios (ref. 2) for the estimation of the committed effective dose equivalent by means of the ingestion of $^{239}Pu+^{240}Pu$ by individuals of different ages. These values are given to crops cultivated in soils with a concentration of $^{239}Pu+^{240}Pu$ of the 2.1×10^{-3} Bq x Kg^{-1} order.

As a result of this finding, the direct ingestion of non - washed tomatoes produced by hectare, would represent as, a maximum, a collective committed effective dose equivalent, $S_{E,70}$, of 3.0×10^{-3} man Sv x $year^{-1}$ for adults. The individual dose would be on the order of 1.5 µSv x $year^{-1}$ when based on a yearly consumption of 40 Kg of tomatoes.

In cultivation of products which are used for feeding domestic animal in soils with this degree of $^{239}Pu+^{240}Pu$ contamination considerably decreases the value of the collective dose, $S_{E,70}$, for the public, since for each cultivated hectare, and according to the production of alfalfa or barley, the collective doses, $S_{E,70}$, would be represented by the consumption of meat by adults, which runs between the range of 0.6×10^{-9} and 7.9×10^{-9} man Sv x $year^{-1}$ and the consumption of milk by babies which runs

between the range of 0.4×10^{-9} and 4.7×10^{-9} man Sv x $year^{-1}$.

In conclusion the cultivation of soils with a less degree of $^{239}Pu+^{240}Pu$ contamination will produce lower values of the collective doses, $S_{E,70}$, in a proportional factor to the relation of concentrations of $^{239}Pu+^{240}Pu$ in the soils. Therefore, the collective dose derived from the cultivated products would be, at least, a 10 factor lower to those expressed in Table 8 in most of the area.

CONCLUSIONS

Field study data obtained through the observation of those crops cultivated in contaminated soils by plutonium, and which were subject to the dilution of superficial contamination by ploughing to a depth of 30-40 cm, as a means of remedial action, allows us to present the following conclusions:

1) In Mediterranean climatological places with scare precipitation, the external surface contamination of those products contaminated by plutonium, by means of resuspension, represents an important process. The values of the soil-crop concentration ratios are in the order of 10^{-4} for tomato fruit and 10^{-3} for the tomato plant and for the diverse components of barley and alfalfa.

2) Dilution by ploughing and homogenization methods used as remedial action in soils with superficial contamination by plutonium on the 120-1200 kBq/m^2 range represents that, for each cultivated hectare for products used as animal feed, the collective committed effective dose equivalent, $S_{E,70}$, proceeding from the beef ingestion of animals fed only with alfalfa or barley (grain and straw) contaminated from these soils, would be, as a maximum, in the order of 10^{-9} man Sv x $year^{-1}$. After the ingestion of milk produced by cows fed with those crops, these would range, as a maximum, in the same order of magnitude, it is to say, about 10^{-9} man Sv x $year^{-1}$ x Ha^{-1}.

This would represent a collective effective dose committment for 10^4 years in the order of 10^{-5} man Sv x Ha^{-1}.

Those products cultivated for human consumption, as tomatoes, for instance, in the zones with this range of contamination by plutonium would represent that for each cultivated hectare, the proceeding $S_{E,70}$ were in the order of 10^{-3} as maximum. However, given the assumptions made from the contamination of the totality of the harvest and the processes to which these products are subject to before their direct ingestion, it would not be a scanty observation to considerer a $S_{E,70}$ in the order of 10^{-4} man Sv x year^{-1}. Therefore, the collective effective dose commitment for 10^{4} years, would be in the order of 1 man Sv by hectare.

The annual committed effective dose equivalent to individuals by the ingestion of tomatoes would be 1.5 μSv.

3) IAEA consider 1 man Sv of collective effective dose equivalent commitment as guideline in stablishing exempt quantities and rules for practical application.

To achieve a collective effective dose commitment no higher than 1 man Sv for 10^{4} years, as a consequence of individual consumption of foodstuffs cultivated in these soils, it has been deduced:

- In those soils devoted to the cultivation of products used as animal feed, ploughing and homogenization methods up to 30 cm depth are sufficient in soils with surface contamination of plutonium of 1200 kBq/m^2.

Therefore, an Intervention Level of 1200 kBq/m^2 would be deduced in order to apply this remedial action to extensions of 10^{4} - 10^{6} hectares devoted to this type of crops.

- In soils devoted to the harvest of products for man's direct ingestion, it is considered that the ploughing and homogenization methods up to a 30 cm depth is sufficient remedial action for the application to soils with a superficial contamination of plutonium of 120 kBq/m^2.

Therefore, an Intervention Level of 120 kBq/m^2 would be the deduced one in order to implement this remedial action to extensions up to 100 hectares devoted to these crops.

The extension of those contaminated areas and the value of the surface contamination will play an important role in the optimization of the Intervention Levels wich could be applied in the case of an accident. Those variations that could emerge in the knowledge of the transference factors and those of the international acceptable norms for determining the value of the collective dose commitment that establishes the exemption limit shoul also be considered.

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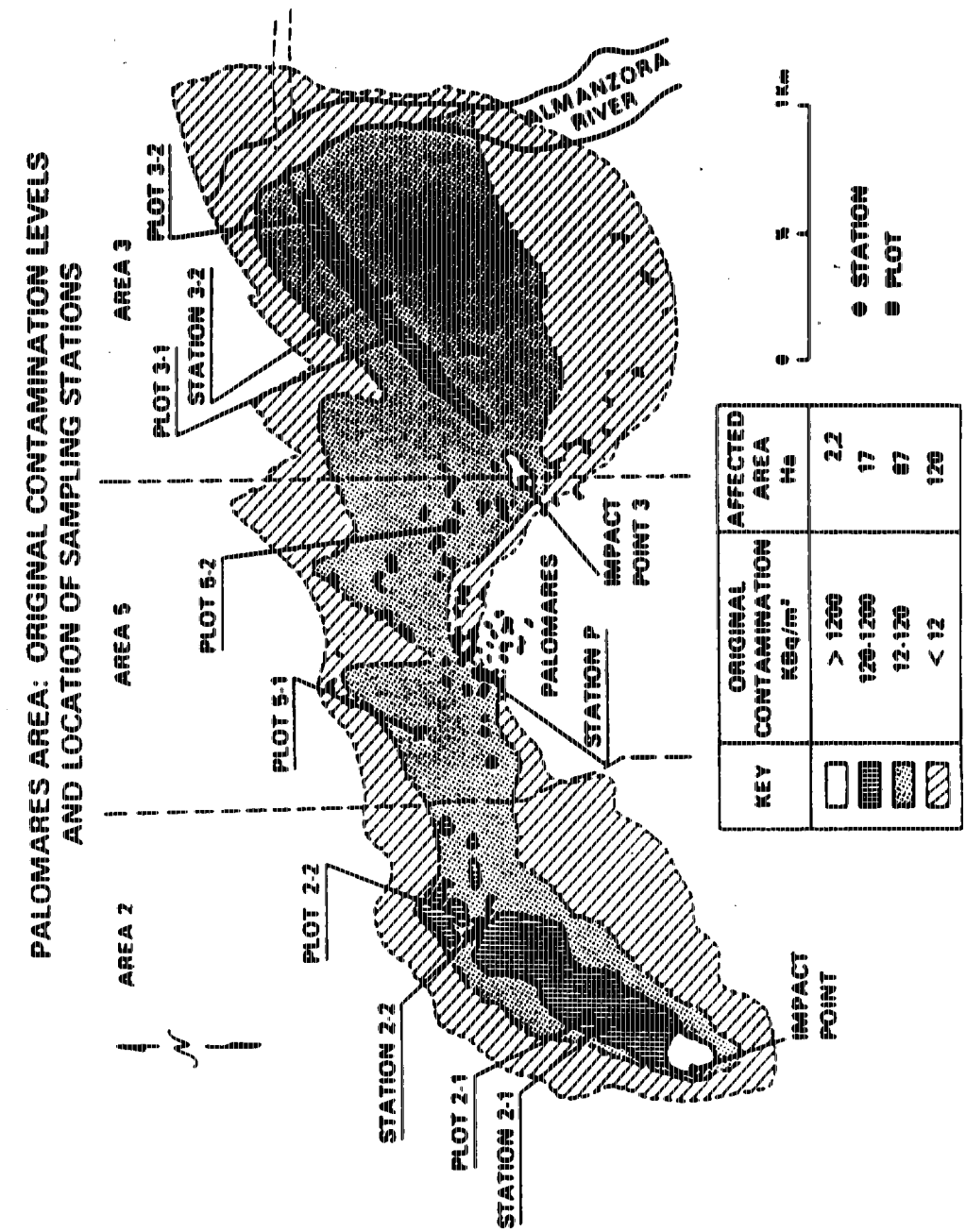
PALOMARES AREA: ORIGINAL CONTAMINATION LEVELS
AND LOCATION OF SAMPLING STATIONS
AREA 2
AREA 5
AREA 3
PLOT 2-2
PLOT 5-1
PLOT 5-2
PLOT 3-1
PLOT 3-2
STATION 2-2
STATION 2-1
STATION 3-2
PLOT 2-1
STATION P
PALOMARES
IMPACT POINT 3
IMPACT POINT
ALMANZORA RIVER
KEY
ORIGINAL CONTAMINATION KBq/m²
AFFECTED AREA Ha
> 1200
1200-120
12-120
< 12
2.2
17
67
120
0
1 Km
STATION
PLOT

REMEDIAL ACTIONS TAKEN AT PALOMARES FOLLOWING THE JANUARY, 1966, ACCIDENT

Location	Hectares*	Acres	Remediation
Impact Point 2	1.6	4	Removed top 10 cm
Impact Point 3	0.6	1.5	Removed top 10 cm
Remainder	224	550	Plowed to 30 cm where possible
Total	226	558	

*259 hectares equal one square mile.

Table 2.- CONCENTRATION OF PLUTONIUN IN STUDY PLOTS.

PLOT	2-1	2-2	3-1	3-2	5-1	5-2
$^{239}Pu + ^{240}Pu$ Concentration $\bar{X}$, K Bq x Kg^{-1}	0.44	2.1	1.1	1.8	0.13	0.29

Table 3.- PLUTONIUM CONCENTRATIONS IN CULTIVATED CROPS FROM PALOMARES.

PLANT		N° SAMPLES		Pu Conc., Bq x Kg^{-1}
Species	Part	Total	% Positive	$\bar{X}$ in positives
TOMATO	Fruit	159	28.3	0.22
TOMATO	Washed fruit	231	6.1	0.15
TOMATO	Plant	206	41.7	4.42
BARLEY	Grain	496	26.8	2.47
BARLEY	Stalk	496	37.0	5.87
BARLEY	Spicule	144	58.3	5.38
ALFALFA	Plant	112	39.0	3.33

Table 4.- PLUTONIUM DISTRIBUTION IN SOILS AND TOMATOES CROPS AND SOIL-PLANT CONCENTRATION RATIOS IN PALOMARES.

Plot	$^{239}Pu + ^{240}Pu$ Concentration in Soil Bq x Kg^{-1}	$^{239}Pu + ^{240}Pu$ Concentration in tomatoes $\bar{X}$, Bq x Kg^{-1}			Soil-plant concentration ratio		
		Fruit	Washed fruit	Plant	Fruit	Washed Fruit	Plant
(2-2)A'	2.1×10^{3}	0.18	0.18	0.56	0.85×10^{-4}	0.85×10^{-4}	0.27×10^{-3}
(2-2)B'	2.1×10^{3}	0.20	0.24	7.68	0.95×10^{-4}	1.14×10^{-4}	3.65×10^{-3}
(2-2)C'	2.1×10^{3}	0.15	0.14	7.20	0.70×10^{-4}	0.70×10^{-4}	3.43×10^{-3}
3-2	1.8×10^{3}	0.45	0.23	3.10	2.50×10^{-4}	1.20×10^{-4}	1.72×10^{-3}
(3-2)A'	1.8×10^{3}	0.28	≤ 0.0004	6.28	1.60×10^{-4}	—	3.49×10^{-3}
3-1	1.1×10^{3}	0.042	0.004	1.79	0.38×10^{-4}	0.04×10^{-4}	1.63×10^{-3}
(3-1)A'	1.1×10^{3}	0.30	0.21	1.21	2.70×10^{-4}	1.90×10^{-4}	1.10×10^{-3}
(3-1)B'	1.1×10^{3}	0.27	0.02	0.04	2.50×10^{-4}	0.18×10^{-4}	0.04×10^{-3}
5-2	0.29×10^{3}	≤ 0.0004	≤ 0.0004	1.52	—	—	5.24×10^{-3}

Table 5.- PLUTONIUM DISTRIBUTION IN SOILS AND BARLEY CROPS AND SOIL-PLANT CONCENTRATION RATIOS IN PALOMARES.

Plot	$^{239}Pu + ^{240}Pu$ Concentration in Soil Bq x Kg^{-1}	^{239}Pu ^{240}Pu Concentration in barley $\bar{X}$, Bq x Kg^{-1}			Soil plant concentration ratio		
		Grain	Straw	Spicule	Grain	Straw	Spicule
2-2	2.1×10^{3}	6.00	8.89	2.16	2.86×10^{-3}	4.23×10^{-3}	1.03×10^{-3}
(2-2) A	2.1×10^{3}	0.82	8.94	8.15	0.39×10^{-3}	4.26×10^{-3}	3.88×10^{-3}
(2-2) B	2.1×10^{3}	0.61	4.82	3.28	0.29×10^{-3}	2.30×10^{-3}	1.56×10^{-3}
3-2	1.8×10^{3}	2.67	3.85	5.92	1.48×10^{-3}	2.14×10^{-3}	3.29×10^{-3}
(3-2)A	1.8×10^{3}	4.65	5.28	6.36	2.53×10^{-3}	2.93×10^{-3}	3.53×10^{-3}
3-1	1.1×10^{3}	1.17	2.72	0.65	1.06×10^{-3}	2.47×10^{-3}	0.59×10^{-3}
(3-1)A	1.1×10^{3}	2.29	12.51	5.48	2.08×10^{-3}	11.37×10^{-3}	4.98×10^{-3}
(3-1)B	1.1×10^{3}	0.99	3.05	10.35	0.90×10^{-3}	2.77×10^{-3}	9.40×10^{-3}
(5-2)A	0.29×10^{3}	0.34	1.36	3.14	1.17×10^{-3}	4.69×10^{-3}	10.83×10^{-3}
(5-2) B	0.29×10^{3}	1.26	1.40	2.60	4.34×10^{-3}	4.82×10^{-3}	8.96×10^{-3}
5-1	0.13×10^{3}	0.43	1.71	2.56	3.31×10^{-3}	13.20×10^{-3}	19.70×10^{-3}

Table 6.- PLUTONIUM DISTRIBUTION IN SOILS AND ALFALFA CROPS AND SOIL-PLANT CONCENTRATION RATIOS IN PALOMARES.

Plot	$^{239}Pu + ^{240}Pu$ Concentration in Soil Bq x Kg	$^{239}Pu + ^{240}Pu$ Concentration in alfalfa $\bar{X}$, Bq x Kg^{-1}	Soil-plant concentration ratio
		Plant	Plant
(2-2)A"	2.1×10^{3}	8.40	4.00×10^{-3}
3-2	1.8×10^{3}	3.20	1.78×10^{-3}
3-1	1.1×10^{3}	40.30	36.60×10^{-3}
5-2	0.29×10^{3}	0.90	3.10×10^{-3}
5-1	0.13×10^{3}	0.98	7.54×10^{-3}
(5-1)A"	0.13×10^{3}	1.06	8.15×10^{-3}
(5-1)B"	0.13×10^{3}	0.66	5.10×10^{-3}
(5-1)C"	0.13×10^{3}	0.68	5.23×10^{-3}

Table 7.- SOIL-PLANT CONCENTRATION RATIOS.

PLANT		CONCENTRATION RATIO
SPECIES	PART	
Tomatoes	Fruit	1.5×10^{-4}
"	Washed fruit	0.9×10^{-4}
"	Plant	2.3×10^{-3}
Barley	Grain	1.9×10^{-3}
"	Straw	5.0×10^{-3}
"	Spicule	6.2×10^{-3}
Alfalfa	Edible	8.9×10^{-3}

Table 8.- COLLECTIVE COMMITTED EFFECTIVE DOSE EQUIVALENT BY INGESTION OF FOOD DERIVED FROM ANNUAL CROPS PER HECTARE CULTIVATED IN SOILS WITH A $^{239}Pu + ^{240}Pu$ CONCENTRATION OF 2.1×10^{3} Bq x Kg^{-1}.

CROP			TOMATOES	BARLEY		ALPHALPHA
				Grain	Straw	
$^{239}Pu + ^{240}Pu$ IN ANNUAL CROP Bq x Ha^{-1} x y^{-1}			25.2×10^{3}	9.98×10^{3}	23.1×10^{3}	130.8×10^{3}
DIRECT CONSUMPTION $S_{E,70}$		Adult	3.02×10^{-3}			
		5 y. old	6.05×10^{-3}			
INDIRECT CONSUMPTION $S_{E,70}$	Beef	Adult		6.0×10^{-10}	1.3×10^{-9}	7.9×10^{-9}
		5y. old		12.0×10^{-10}	2.7×10^{-9}	15.7×10^{-9}
	Milk	Adult		1.2×10^{-10}	2.8×10^{-10}	1.6×10^{-9}
		1y. old		3.6×10^{-10}	8.32×10^{-10}	4.7×10^{-9}

GEOCHEMICAL DISTRIBUTION OF PLUTONIUM AND AMERICIUM IN PALOMARES SOIL

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GEOCHEMICAL DISTRIBUTION OF PLUTONIUM AND AMERICIUM IN PALOMARES SOIL

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Abstract. An aerial accident on January 17, 1966, over the Palomares (Almeria) region, caused four thermonuclear bombs to be dropped, resulting in fractionation of two of them and ignition of part of their core components. The aerosol created by this accident contaminated 226 hectares of cultivated and unimproved calcareous-silicic land.

Soil samples from some chosen parcels were sampled fifteen years after the accident to study the geochemical distribution of plutonium and americium. The soil in each of the parcels of land has been divided into nine parts in order to analyze the distribution of actinides from particles of less than 5 microns to those of more than 1 mm in size, with the maximum build-up falling between 63 and 250 microns. In each of these divisions studies have been made of the checmical and mineral composition of the soil, and through radiographic techniques it has been determined that the minerals most responsible for actinide retention are iron oxides, carbonates and phillosilicates.

In addition, studies were made to determine the electrochemical potential and chemical composition of equilibrium waters obtained by maceration of soils from each parcel. Values for these potentials are between 7.6 and 8.4 for pH, between 312 and 344 mV for Eh and between 484 and 2500 μS for conductivity. The salinity usually found is partially conditioned by the brackish waters used for irrigation.

1. INTRODUCTION

On January 17, 1966, an aviation accident took place during a supply flight over the Palomares airspace in the municipality of Cuevas de Almanzora, province of Almeria. This accident resulted in four thermonuclear bombs being dropped; two of them had parachutes and were recovered intact, one in the dry riverbed of the mouth of the Almanzora River, the other in the sea. The other two bombs fell without parachutes and suffered fragmentation of fissionable fuel with ignition of part of it. The resulting aerosol was carried by the wind and contaminated, to differing degrees, an area of 226 hectares comprising brushlands, farmland and urban areas. Most of the urban area was not contaminated.

Figure 1 shows the contaminated regions defined according to the degrees of superficial alpha contamination measured immediately after the accident.

In places where superficial alpha contamination reached values exceeding 1.2 MBq x m^{-2}, the surface layer was removed to a depth of approximately 10 cm. This layer of soil and contaminated vegetation was provisionally deposited in a site near impact number 2, packed in steel drums and later sent to the United States as radioactive wastes. The rest of the contaminated region used for agriculture was watered and plowed to a depth of approximately 30 cm in order to dilute the contaminated soil with clean soil and thus reduce the concentration of radionuclides on the surface.

This report deals with the studies that have been made to determine levels of correlation existing between Pu-239 + Pu-240 and Am-241 concentration and the granulometric, mineralogical and chemical composition of the soil. These aspects are important for evaluating the risks attending dispersion of such contaminants through atmospheric re-suspension, erosion and hydrological movement.

2. METHODOLOGY OF SAMPLING AND ANALYSIS

2.1. Sampling

The samples used in this study were taken from parcels 2-0, 2-2, 3-1, 3-2, and 5-3B. Their location is indicated in Figure 1.

Parcels 2-2, 3-1, 3-2, and 5-3B are usually cultivated; parcel 2-0 has been under cultivation for only one year. Parcel 5-3B is situated outside the area that was initially contaminated.

The samples were taken in 1981 from regular points distributed along diagonals on a squared area (50 x 50 m) of each parcel, to a depth of 15 cm.

2.2. Analyses

Analyses were made of aliquote parts obtained through splitting dry samples by means of a Centrifugal Sample Divider.

2.2.1. Granulometry

The samples were subjected to maceration and ultrasonic separation. From each sample, eight size fractions were obtained with the following size intervals: $\geqslant$ 1000-250-63-40-20-10-5 $\leqslant$ µm.

Fractional division of $>$ 63 µm particles, after leaching with water over a mesh of a prescribed size, was carried out by screening according to NLT 104/72 standards (1).

Particles $<$ 63 µm in size were fractionated by the "British Rema" air classifier; mean density was considered to be 2.7 gm/cm^3.

Granulometric distribution of components $<$ 5 µm in size was found with

a "Coulter TA II" particle counter.

Sherard methodology was used to determine the dispersion index (2).

2.2.2. Americium

The presence of Am-241 was measured by analysis of 15 gr samples through low-energy photon spectrometry with intrinsic germanium detectors.

2.2.3. Plutonium

The measurement of Pu-239 and Pu-240 was carried out on the same samples as were proviously used for measuring Am-241, by alpha spectrometry using silicon block semiconductor detectors. The process calls for prior acid extraction with 1M HF and 12 N HNO_3, separation of plutonium with DOWEX AG-1X2 (50-100 mesh) resin, and electrodeposit of the evaporated resin eluate residue dissolved in HCl and ammonium oxalate. The chemical yield is measured by addition of Pu-236 as a tracer.

2.2.4. Mineralogical Composition

The mineralogical analysis was carried out through optic microscopy and X.R diffraction. The second technique was used to analyze each of the granulometric fractions. The quantitative analysis of the mineral species was based on the intensity of the most important reflections. The value of the intensities, I, represents arbitrary units, being 100 the value of I for the highest reflection intensity in the different granulometric fractions.

2.2.5. Autoradiographies

They were obtained on plastic, type CR-39 (oxydi-2, 1-ethanedyl di-2-

propenyl diester of carbonic acid), placed over thin polished sections of the granulometric fractions during 32 days.

2.2.6. Chemical Composition and Physico-chemical Parameters

The cationic and anionic macroconstituents were determined on solid sample and in water obtained from maceration and leaching of each of the samples. The techniques used were: Emission spectrometry with plasma source, ionic cromatography and specific ion electrode, turbidimetry, C-S analyzer and fluorimetry.

The Eh, pH and conductivity were determined from soil elutriation waters.

3. RESULTS AND DISCUSSION

The results of the activity concentration of ^{239}Pu + ^{240}Pu and ^{241}Am in the granulometric fractions corresponding to the soil of the selected parcels and their correlation with the mineralogic and chemical composition of those soils are discussed in the following paragraphs.

3.1. Pu and Am concentration

Table I shows the concentration and the activity ratios of ^{239}Pu + ^{240}Pu and ^{241}Am in each of the parcels, obtained as the mean value of all the measures carried out in the granulometric fractions. From these values we can deduce:

- The existence of a notable difference between the contamination levels of parcels 2 and the other lots.

- The Pu/Am ratio in samples from zone 2-2, with values > 6, and from zones 3-2 and 5-3B seem to indicate that a fractioning due to a higher

leaching of Am has occured, given that the theoretical ratio calculated from the desintegration of ^{241}Pu content in 1966 was 3.05. In lots, 2-0 and 3-1, the experimental ratio is more or less similar to the theoretical one.

- The Pu/Am ratio is almost constant in the different size fractions, with a coefficient of correlation above 0.98 except in the 5-3B sample where no correlation is found. Nevertheless, Table II shows that in the very small sizes (≤ 10 µm) this ratio always increases.

Tables III and IV include the activity concentrations of Pu and Am for each of the size fractions. These values indicate that:

- Plutonium and americium concentration is maximal in the fractions from 63 to 250 µm, except in sample 5-3B where maximum concentration occurs in the portion smaller than 5 µm. A second maximum concentration in the 20 to 40 µm group was found in samples 3-1 and 3-2.

- Concentration of plutonium and americium in fractions larger than 250 µm is much lower that for all the remaining fractions and almost negligible in comparison.

- In fractions smaller than 250 µm, the coefficient of correlation between both actinides and the size of each fraction is greater than +0.75 except in sample 5-3B where it is -0,5.

Tables V and VI show the distribution (per cent) of the plutonium and americium content of the soil. From this it is deduced that:

- In the parcels of land contaminated by the accident, concentrations of Pu-239 + Pu-240 and Am-241 activity associated with particles smaller that 10 µm are never greater than 15% of existing activity concentration in the soil; in parcels 3-1 and 3-2 (heavy cultivation) it is greater than 10% and in parcels 2-0 and 2-2 it is less than 10%.

In the most heavily cultivated lands, the activity associated with

fractions smaller than 5 μm is 8.74% maximum, while the less cultivated lands show a maximum value of 2.74%.

- For both Pu-239 + Pu-240 and Am-241 the percentages of activity found in fractions from 40 to 250 μm are greater than 75% in parcels 2-0, 2-2, and 3-2. In parcel 3-1, activity in the same size group comes to only about 50%, since an important proportion, about 30%, is found in the particles between 20 and 40 μm in size.

It is in the range from 63 to 250 μm where the greatest proportion is found, accounting for over 50% of total activity in the soil.

- In parcel 5-3B, where contamination has been the consequence of dispersion of contaminants during the 15 years that have elapsed since the accident, 57% of Pu-239 + Pu-240 activity in the soil is found in fractions smaller than 5 μm, while the same group shows only 7% of Am-241 activity.

In fractions between 40 and 250 μm only 9.83% of plutonium and 16.27% of americium are found.

- The percentage of Pu-239 + Pu-240 and Am-241 activity found in fractions larger than 250 μm is insignificant compared with the other fractions.

- An effort was made to obtain an idea of the distribution of plutonium and americium in particles smaller than 5 μm: using sample 2-0 the concentration of plutonium was estimated on the basis of α activity in fractions obtained through sieve analysis by sedimentation, assuming the aforementioned proportional distribution. The results are shown on Table VII and it can be deduced that only 0,4% of all the plutonium contained in the sample from parcel 2-0 is contained in fractions smaller than 0,6 μm.

3.2. Mineralogical Composition

The samples that were studied are composed of silty sand. The propor-

tion of sands ($t \geq 63$ μm) predominates with about 48% against 38% of silt (63 μm $\geq t \geq 5$ μm) and 14% of "clays" (5 μm $\geq t$).

Tables VIII and IX show the granulometric distributions of the total sample and the < 5 μm fraction respectively. Fig. 2A shows the accumulated granulometric distribution of the samples and Fig. 2B shows the accumulated distribution of plutonium. From these figures, the marked relation between size and plutonium content becomes very evident.

According to the petrographic composition, the samples are classified as lithograywacke-phylloarenite (3).

Mean composition, estimated by optic microscope is: 40% quartz, 29% muscovite-illite, 2% chlorite, 22% carbonates (calcite and dolomite), 3% iron oxides and opaque minerals, and 2% vegetal remains. These components represent more than 95% of the total composition.

Other components, estimated at less than 1%, are: garnet, plagioclase, hornblende, pyroxene, epidote, zoisite, tourmaline, zircon, rutile and apatite.

X-ray diffraction study of the mineralogical composition of each of the granulometric fractions are shown in Tables X to XIV.

The general tendency of variations in content in the size fractions, for each of the mineral components, is the following:

- Chlorite and muscovite-illite: irregular distribution, maximum values between 20 and 250 um.

- Quartz: its content tends to diminish with grain size.

- Calcite: distribution reflects both textural forms; sparite: decreases as sizes approach 63 μm; micrite: increases from 63 μm down to the finest sizes.

- Dolomite: irregular distribution; most frequent maximums found in sizes over 40 μm

In the study of the association between minerals, it was deduced that the most significant values for the linear coefficient of correlation were +0.71 for chlorite-moscovite and -0.64 for quartz-dolomite.

The coefficients of correlation between mineral components and plutonium are listed in Table XV.

According to these coefficients, plutonium will be in some way associated with the muscovite-illite in samples 2-0, 3-1 and 3-2; with dolomite in samples 2-0 and 2-2; and with chlorite in sample 3-1. In sample 5-3B plutonium would be independent of the mineralogical composition, tending to an association with calcite.

Coefficients of correlation between americium and mineral components are analogous.

Fig. 3 present the concentration distributions of plutonium and americium and mineralogical proportions in the granulometric fractions of the sample corresponding to parcel 2-0. Here, variations in distribution and the existing correlation between the distribution of plutonium and americium and the distribution of muscovite-illite and dolomite can be seen in more detail.

The autoradiographic analyses yielded the findings seen in the photomicrographs presented in Figures 4 and 5. The results can be summarized as follows:

- There is an association between mineral components and plutonium and americium content of the soil.

- The main concentrations of actinides, estimated from the density of α traces, are found in some fragments of iron oxi-hydroxide; a lesser concentration is found in some carbonate fragments and weak concentrations

in micaceous aggregates. In all of the components indicated, there are fragments with high relative concentrations and others with weak or nil density of traces, but microscopic analysis did not reveal significant genetic differences among them.

- In the fine fractions (≤ 63 µm), high concentration impacts have been recorded without being able to identify the mineral support. The relation between the impact size and effective hydraulic size in the granulometric separation seems to make it necessary to discard the hypothesis of the existence of isolated plutonium oxide particles.

- Vegetal remains have been found with some actinide concentration.

3.3. Chemical Composition and Physical-Chemical Parameters

Tables XVI and XVII show the results of chemical analyses of the samples.

By chemical composition, the samples are classified within the graywacke-lithic group because of their SiO_2 and Al_2O_3 content, although K_2O and Na_2O contents are the inverse of the mean for this type of rock (3).

Then mean content of organic carbon, 0.27%±0.03 can be considered to be within the limits of the graywacke sediment type and lower than the average values of soils that can support some kind of vegetation. Humic acids represent 47% of total organic carbon.

An analysis of maceration-elutriation waters is found in Table XVIII and values for Eh, pH and conductivity are included in Table XIX. In keeping with these results, the water from samples 3-1, 3-2, and 5-3B belongs to the chloro-sulphate group, that of 2-0 to the bicarbonate group, and that of 2-2 to the chloride group, according to their positions on the saline anionic classification diagram (4). From the point of view of cationic content, the water of samples 2-0 and 2-2 is sodic-potassic; that of 3-2 and 5-3B is calcic; and in 3-1 the relative proportions of Na+K, Ca and Mg cations are practically the same.

The total salinity and ionic content of sample 2-0 are significantly different than those of samples 3-1, 3-2, and 5-3B. This difference is probably due to the brackish character of the waters used for irrigation of the cultivated parcels. In parcel 2-2 the effects of cultivation and irrigation are less pronounced.

The Eh and pH values of the samples indicate an oxidizing and slightly alkaline environment. Eh varies from 312 to 344 mV and pH from 7.6 to 8.43. Figure 6 shows the region in which the samples are situated on the Eh-pH diagram in regard to plutonium ion species in solution, in equilibrium with $PuO_{2(s)}$ at 25ºC and 1 atmosphere (5).

According to the Eh-pH diagram it would seem that we are in the theoretical region of stability of PuO_2, although the equilibrium concentration with PuO_2^+ shown in the above-mentioned concentration values is higher than the theoretical value. This seems to indicate the presence of other chemical species.

The content of plutonium in waters and the percentage leached during maceration and elutriation are shown in Table XX. The values for plutonium leaching are a function of the Eh and pH potentials. Greater leaching is achieved in the samples that are most oxidized and lower in pH.

4. CONCLUSIONS

The following are the most significant deductions drawn from the material presented above:

1st. Plutonium and americium content and soil granulometry are directly associated (coefficient of correlation = 0.75). The maximum concentrations correspond to fractions ranging between 63 and 250 µm. Less than 15% of residual plutonium and americium contamination is associated with the fractions of particles smaller than 10 µm. The maximum Pu-239 + Pu-240 found in the portion smaller than 5 µm corresponds to 9% in heavily

cultivated soil and 3% in less cultivated soil. The maximum percentages for Am-241 in the same group smaller than 5 μm are analogous in less cultivated soil and somewhat lower in the heavily cultivated soil.

In parcel 5-3B, which was contaminated well after the accident, Pu-239 + Pu-240 activity in the fraction smaller than 5 μm was 57%.

2nd. The contaminants, plutonium and americium, have been found in some authigenetic iron oxides, carbonates (dolomite) and muscovite-illite. The maximum concentrations were found in the iron oxi-hydroxides.

3rd. The plutonium-americium ratio calculated for the different size fractions has a mean value of 3.0 ± 0.2 in parcels 2.0 and 3.1, while for parcel 2-2 it is 6.7 ± 0.5 and for parcel 3-2 it is 7.4 ± 0.6. This would seem to indicate greater mobilization of americium in the latter three parcels which could probably be related to the Eh and pH potentials of the waters in "equilibrium" with the soil, which in parcels 2-2 and 3-2 create a slightly more reducing and alkaline environment.

4th. The potential leaching of plutonium estimated from the concentrations measured in the maceration-elutriation waters is found in the range of 2×10^{-4} 21×10^{-4} per cent of the total content in the soil, with the greater values corresponding to the most oxidized and least alkaline soils.

5th. It is necessary to continue research aimed mainly at defining the mechanisms of plutonium and americium fixing and dispersion since even though the dispersion factor of clays in these soils is very low (0.23), mobilization of plutonium and americium colloids and complexes remains unknown.

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TABLE I. MEAN CONCENTRATION OF Pu AND Am IN THE SAMPLES

Radionuclide	Activity Concentration (B_q/g)				
	Parcels				
	2-0	2-2	3-1	3-2	5-3B
^{239}Pu ^{240}Pu	42.60	15.60	1.09	1.82	0.46
^{241}Am	13.86	2.54	0.34	0.32	0.02
Ratio Pu/Am	3.0±0.2	6.7±0.5	3.0±0.2	7.4±0.6	41.9±27.8

TABLE II. ACTIVITY CONCENTRATION RATIO OF $^{239}Pu+^{240}Pu/^{241}Am$ IN SIZE FRACTIONS OF SOILS

Parcel \ Fraction µm	≤ 5	5-10	10-20	20-40	40-63	63-250	250-1000
2-0	2.99	3.90	3.22	2.21	2.93	3.21	2.80
2-2	9.21	5.83	5.08	6.14	6.09	6.10	8.33
3-1	4.08	3.28	2.05	3.39	2.29	3.18	2.85
3-2	8.60	9.33	7.38	6.08	6.50	5.22	8.89
5-3B	222	15.67	8.71	9.50	16.67	12.86	7.78

TABLE III. ACTIVITY CONCENTRATION OF $^{239}Pu+^{240}Pu$ IN SIZE FRACTIONS OF SOILS (Bq/g)

Parcel \ Fraction µm	≤ 5	5-10	10-20	20-40	40-63	63-250	250-1000
2-0	8.55	24.57	19.83	15.48	87.28	97.13	0.98
2-2	3.13	3.09	4.68	9.33	29.16	49.88	0.05
3-1	0.53	0.59	0.43	2.24	0.80	2.54	0.02
3-2	0.86	1.12	0.59	1.46	0.39	5.48	0.08
5-3B	2.22	0.47	0.61	0.57	0.15	0.09	0.07

TABLE IV. ACTIVITY CONCENTRATION OF ^{241}Am IN SIZE FRACTIONS OF SOILS, (Bq/g)

Parcel \ Fraction μm	< 5	5-10	10-20	20-40	40-63	63-250	250-1000
2-0	2.86	6.30	6.15	7.00	29.77	30.26	0.35
2-2	0.34	0.53	0.92	1.52	4.79	8.18	0.00
3-1	0.13	0.18	0.21	0.66	0.35	0.80	0.007
3-2	0.10	0.12	0.08	0.21	0.06	1.05	0.009
5-3B	0.01	0.03	0.07	0.06	0.009	0.007	0.009

TABLE V. PERCENTAGE DISTRIBUTION OF $^{239}Pu+^{240}Pu$ IN SIZE FRACTIONS OF SOILS

Parcel \ Fraction μm	< 5	5-10	10-20	20-40	40-63	63-250	250-1000
2-0	2.74	6.07	3.32	7.25	21.25	59.30	0.07
2-2	2.57	1.53	1.32	7.04	18.18	59.33	0.03
3-1	8.74	6.12	2.12	31.79	3.68	47.22	0.33
3-2	7.02	4.55	5.31	7.08	0.71	74.32	0.51
5-3B	56.98	6.05	5.62	20.30	2.88	6.95	1.22

TABLE VI. PER CENT DISTRIBUTION OF ^{241}Am IN SIZE FRACTIONS OF SOILS

Parcel \ Fraction μm	< 5	5-10	10-20	20-40	40-63	63-250	250-1000
2-0	2.82	4.79	3.17	10.07	22.28	56.80	0.07
2-2	1.68	1.61	1.59	7.03	18.32	69.75	0.02
3-1	6.82	5.97	3.34	30.17	5.25	47.94	0.51
3-2	4.55	2.78	3.95	6.46	0.60	81.34	0.32
5-3B	7.02	8.26	14.38	50.46	3.86	12.41	3.61

TABLE VII. DISTRIBUTION OF THE TOTAL α ACTIVITY IN FRACTIONS < 5 µm OF SAMPLE 2-0

µm	Percentage	Activity Concentration (Bq/g)
5.00 - 2.10	6.70	10.59
2.10 - 1.80	0.76	2.84
1.80 - 0.87	1.27	18.17
0.87 - 0.79	0.28	16.07
0.79 - 0.60	0.54	4.13
0.60	4.13	4.00

TABLE VIII. PER CENT GRANULOMETRIC DISTRIBUTION OF THE SOILS

Parcel \ Fraction µm	<5	5-10	10-20	20-40	40-63	63-250	250-1000	>1000
2-0	13.68	10.53	7.14	19.95	10.37	26.01	2.99	9.33
2-2	12.76	7.72	4.40	11.75	9.71	21.63	9.26	22.77
3-1	17.94	11.36	5.32	15.41	5.02	20.15	20.02	4.78
3-2	14.91	7.39	16.46	8.82	3.33	24.80	11.86	12.43
5-3B	11.80	5.86	4.20	16.23	8.81	35.85	8.65	8.90

TABLE IX. PER CENT GRANULOMETRIC DISTRIBUTION OF THE < 5 µm FRACTION OF SOILS

Parcel \ Fraction µm	1.0-1.3	1.3-1.6	1.6-2.0	2.0-2.6	2.6-3.2	3.2-4.1	4.1-5.1	5.1-6.5
2-0	7.57	9.34	10.87	13.11	15.65	15.53	15.20	12.73
2-2	13.91	14.01	17.55	14.13	10.93	10.04	10.16	9.27
3-1	28.81	17.52	7.02	7.66	9.53	10.07	10.62	8.77
3-2	26.95	10.42	7.82	8.50	10.98	11.66	12.59	11.08
5-3B	33.85	19.07	6.92	6.55	8.22	8.11	8.70	8.58

TABLE X. MINERALOGICAL COMPOSITION OF SIZE FRACTIONS

	Parcel 2-0				
I / Size µm	Chlorite d(Å)-14	Moscovite-Illite d(Å)-10	Quartz d(Å)-4.24	Calcite d(Å)-3.03	Dolomite d(Å)-2.88
1000	100.00	50.70	49.89	69.35	20.48
250	41.30	33.80	100.00	73.99	19.58
63	80.43	93.43	84.92	49.54	54.81
40	62.22	100.00	58.09	39.00	100.00
20	84.78	97.18	55.43	65.33	25.90
10	58.70	59.62	21.51	61.30	7.53
5	58.70	72.24	11.97	76.78	10.54
	67.39	46.48	6.65	100.00	5.42

TABLE XI. MINERALOGICAL DISTRIBUTION OF SIZE FRACTIONS

	Parcel 2-2				
I / Size µm	Chlorite d(Å)-14	Moscovite-Illite d(Å)-10	Quartz d(Å)-4.24	Calcite d(Å)-3.03	Dolomite d(Å)-2.88
1000	23.73	24.40	71.52	53.63	86.36
250	33.90	32.37	100.00	38.64	50.91
63	44.07	35.75	97.43	14.52	100.00
40	38.98	92.75	57.60	43.56	51.14
20	100.00	100.00	59.10	63.70	68.18
10	61.02	93.72	20.77	61.59	57.95
5	27.12	33.09	13.06	79.16	39.77
	37.29	31.64	5.78	100.00	--

I = reflexion intensity in arbitrary units, taking 100 as the maximum value of I of the mineral in the X-Ray difraction diagrams.

TABLE XII. MINERALOGICAL DISTRIBUTION OF SIZE FRACTIONS

	Parcel 3-1				
I / Size µm	Chlorite d(Å)-14	Moscovite-Illite d(Å)-10	Quartz d(Å)-4.24	Calcite d(Å)-3.03	Dolomite d(Å)-2.88
1000	3.89	22.55	85.96	58.37	55.23
250	5.63	29.30	100.00	53.47	45.35
63	100.00	100.00	67.98	66.12	41.86
40	6.36	55.32	64.45	58.18	100.00
20	5.65	90.21	35.09	58.78	29.65
10	9.89	78.30	18.20	71.43	19.19
5	13.78	85.74	11.84	95.51	27.33
	14.19	64.26	8.11	100.00	11.63

TABLE XIII. MINERALOGICAL COMPOSITION OF SIZE FRACTIONS

	Parcel 3-2				
I / Size µm	Chlorite d(Å)-14	Moscovite-Illite d(Å)-10	Quartz d(Å)-4.24	Calcite d(Å)-3.03	Dolomite d(Å)-2.88
1000	27.59	58.26	99.77	62.00	16.15
250	31.03	39.99	79.25	57.20	85.84
63	81.03	99.08	75.76	82.00	50.66
40	24.14	42.20	100.00	56.80	100.00
20	93.10	100.00	56.88	75.20	40.27
10	34.48	66.51	30.30	53.60	10.62
5	100.00	91.74	14.92	85.20	7.96
	58.62	64.68	8.39	100.00	4.42

TABLE XIV. MINERALOGICAL COMPOSITION OF SIZE FRACTIONS

I / Size µm	Parcel 5-3B				
	Chlorite d(Å)-14	Moscovite-Illite d(Å)-10	Quartz d(Å)-4.24	Calcite d(Å)-3.03	Dolomite d(Å)-2.88
1000	53.49	40.51	62.27	100.00	30.00
250	51.16	58.56	89.64	65.24	92.50
63	88.37	83.33	97.16	35.24	46.67
40	27.91	46.30	100.00	57.14	90.00
20	100.00	99.07	77.29	47.38	71.67
10	90.70	100.00	26.84	55.24	26.67
5	79.07	81.48	15.03	62.38	21.67
	69.77	74.07	21.74	71.90	10.00

TABLE XV. LINEAR COEFFICIENT OF CORRELATION BETWEEN THE ACTIVITY CONCENTRATION OF $Pu^{239+240}$ AND THE MINERALS

ρ / Parcel	Correlated Parameters: Activity Concentration of $^{239}Pu + ^{240}Pu$ Versus Mineral				
	Chlorite	Moscovite-Illite	Quartz	Calcite	Dolomite
2-0	0.39	**0.73**	0.33	**-0.78**	**0.84**
2-2	-0.04	0.03	**0.51**	**-0.72**	**0.78**
3-1	**0.66**	**0.75**	0.06	-0.25	0.02
3-2	**0.50**	**0.66**	0.02	0.37	-0.06
5-3B	0.15	0.16	**-0.64**	**0.55**	0.31

TABLE XVI. CHEMICAL COMPOSITION PER CENT OF MACROCONSTITUENTS IN SAMPLES OF SOILS

TOTAL SAMPLE

Oxides \ Parcels	2-0	2-2	3-1	3-2	5-3B
*SiO_2	53.00	58.00	53.00	55.00	56.00
Al_2O_3	12,10	8.20	13.30	12.20	9.00
CaO	10.60	11.80	9.40	9.00	10.80
MgO	2.10	1.60	2.20	2.20	2.00
MnO	0.05	0.06	0.09	0.07	0.06
Fe_2O_3	4.70	3.60	5.50	5.10	4.30
Na_2O	0.86	0.53	0.96	0.85	0.63
K_2O	2.60	1.50	2.70	2.60	1.90
TiO_2	0.60	0.47	0.56	0.55	0.51
P_2O_5	0.16	0.09	0.19	0.19	0.12
**P.C.	13.00	14.00	12.00	12.00	15.00

*). SiO_2 calculated by difference to 100.

**). P.C.: Calcination losts at 900ºC during 2 hrs.

TABLE XVII. ADDITIONAL SIGNIFICANT COMPONENTS FROM SAMPLES OF SOILS

TOTAL SAMPLE

Components \ Parcel	2-0	2-2	3-1	3-2	5-3B
% Total C	3.030	3.730	2.740	2.680	3.560
% Org. C	0.389	0.321	0.226	0.192	0.210
% Total S	0.011	0.022	0.279	0.170	0.247
U_3O_8 ppm	4.000	4.000	3.000	4.000	4.000
Humic acids ppm	1710	1590	1166	917	1545

TABLE XVIII. ACTIVITY ($mol.l^{-1}.10^{-3}$) AND IONIC STRENGTH CALCULATED IN MACERATION-ELUTRIATION WATEPS FROM SAMPLES OF SOILS

Ions \ Parcels	2-0	2-2	3-1	3-2	5-3B
HCO_3^-	2.54	4.56	1.43	1.53	1.04
Cl^-	0.48	10.71	15.67	9.49	30.24
NO_3^-	0.07	0.06	3.54	1.01	2.68
NO_2^-	0.03	0.01	0.41	0.34	0.12
PO_4^{3-}	-	0.01	-	-	-
$SO_4^=$	0.59	0.72	6.74	2.47	8.77
Na^+	2.25	10.94	10.67	7.44	10.44
K^+	1.71	1.61	1.93	1.09	2.03
Ca^{++}	0.64	0.50	3.07	5.49	4.94
Mg^{2+}	0.25	0.79	3.88	2.30	2.04
Ionic Strength	8.37	24.18	89.99	57.16	118.81

TABLE XIX. PHYSICAL-CHEMICAL PARAMETERS OF THE SYSTEM WATER/SOIL FROM THE SAMPLES OF SOIL

Parcels \ Parameters	Eh (mV)	pH	Conductivity µS
2-0	340.4	7.70	484
2-2	312.0	8.20	1247
3-1	344.4	7.60	2500
3-2	328.4	8.43	2330
5-3B	321.4	8.36	2410

TABLE XX. PLUTONIUM CONCENTRATION IN MACERATION-ELUTRIATION WATERS

Parcels	PuO_2, ($Mol.l^{-1}.10^{-15}$)	% leached
2-0	250	12×10^{-4}
2-2	10	2×10^{-4}
3-1	10	21×10^{-4}
3-2	5	5×10^{-4}
5-3B	1	5×10^{-4}

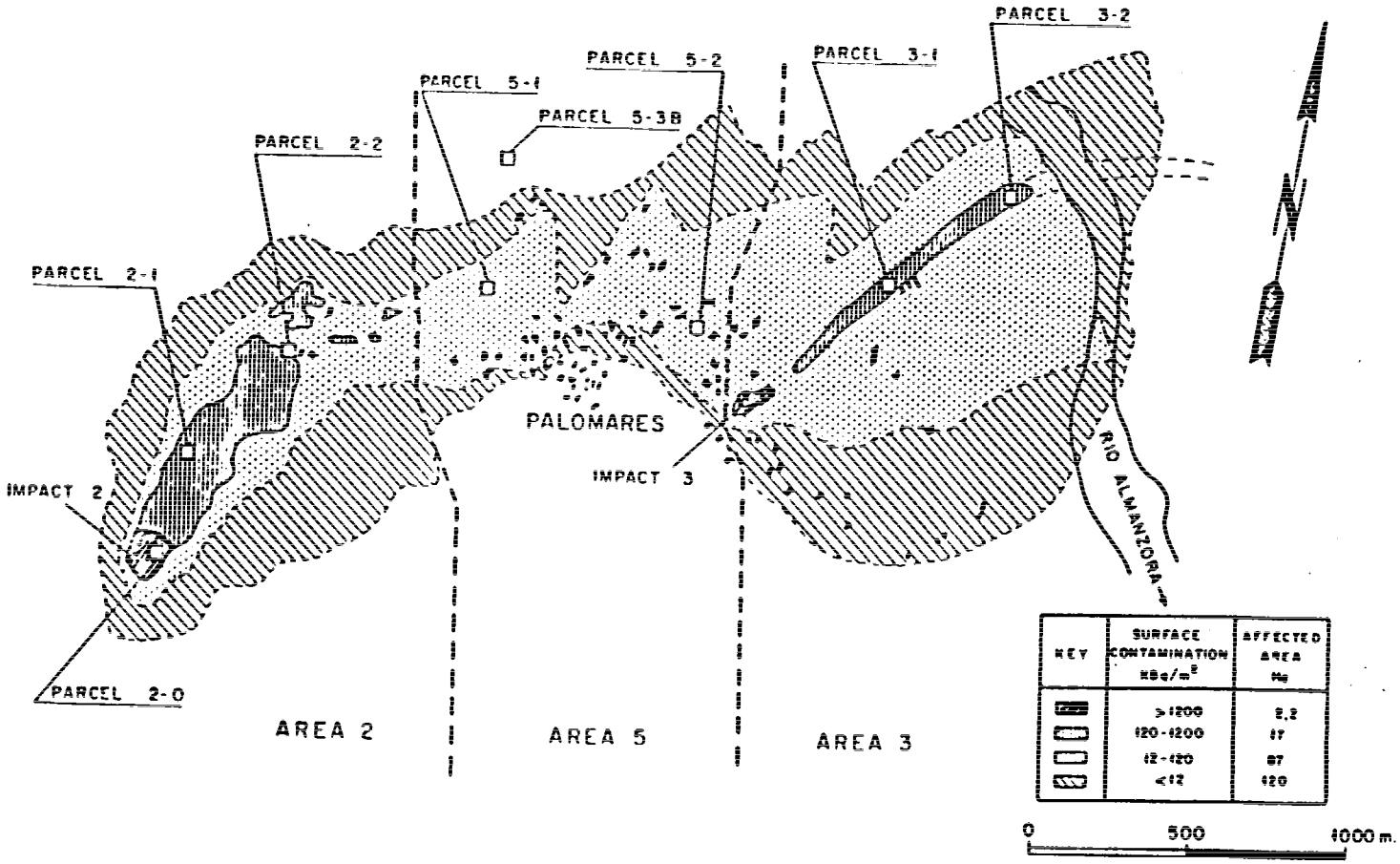


FIG. 1.- PALOMARES REGION

LEVELS OF ORIGINAL CONTAMINATION AND LOCATION OF SAMPLE PARCELS

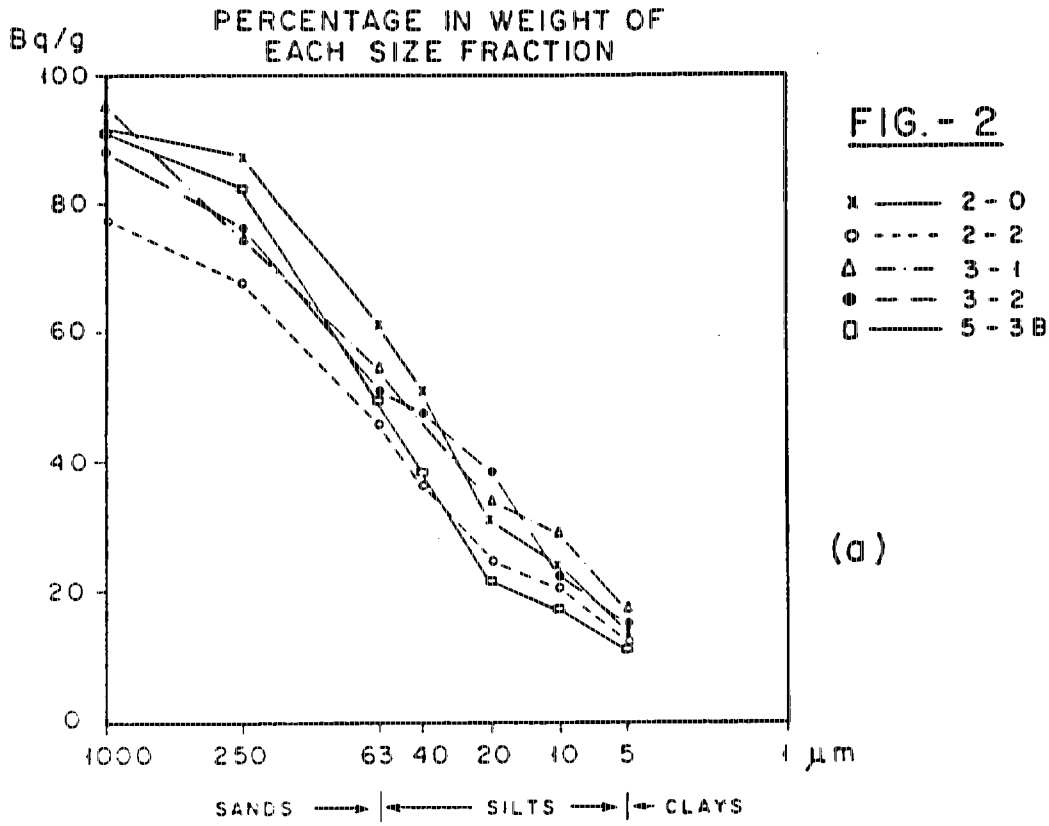
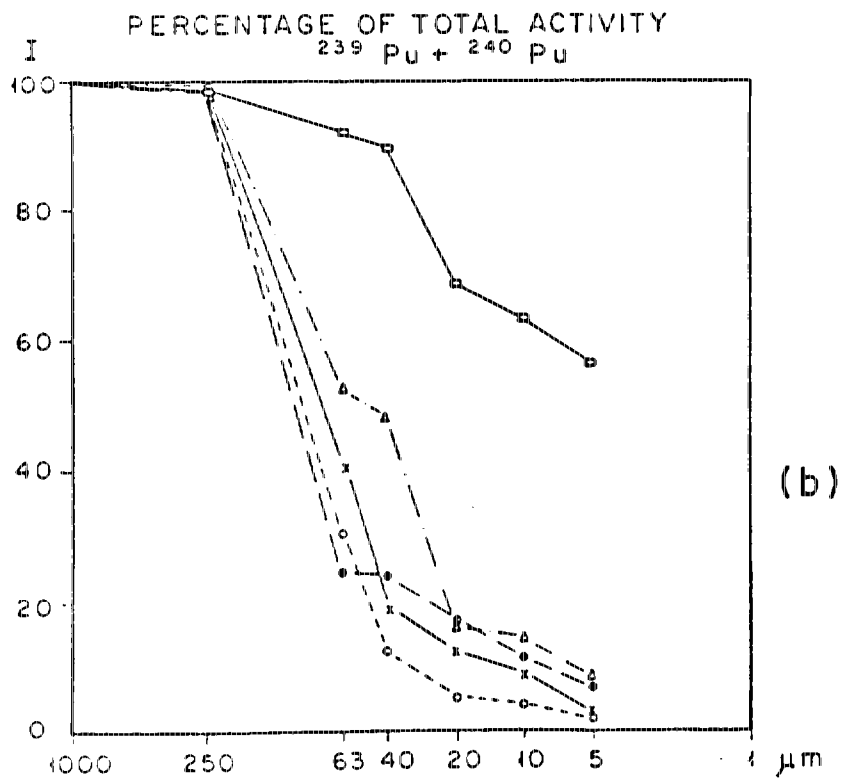

PERCENTAGE IN WEIGHT OF EACH SIZE FRACTION
Bq/g
100
80
60
40
20
0
1000
250
63
40
20
10
5
µm
SANDS
SILTS
CLAYS
x —— 2 - 0
o ---- 2 - 2
Δ —·— 3 - 1
● — — 3 - 2
□ —— 5 - 3B
(a)

FIG. - 2


PERCENTAGE OF TOTAL ACTIVITY
239 Pu + 240 Pu
I
100
80
60
40
20
0
1000
250
63
40
20
10
5
µm
(b)

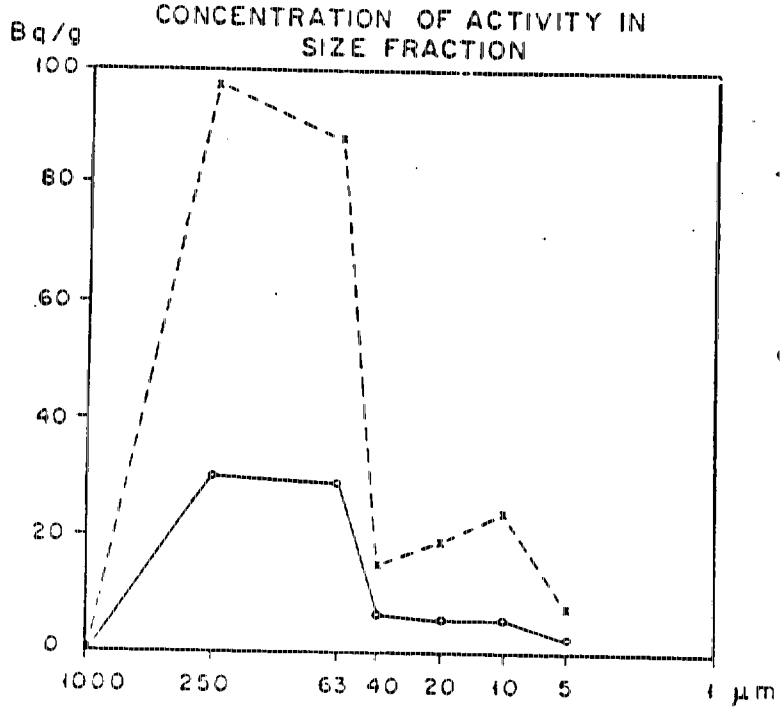
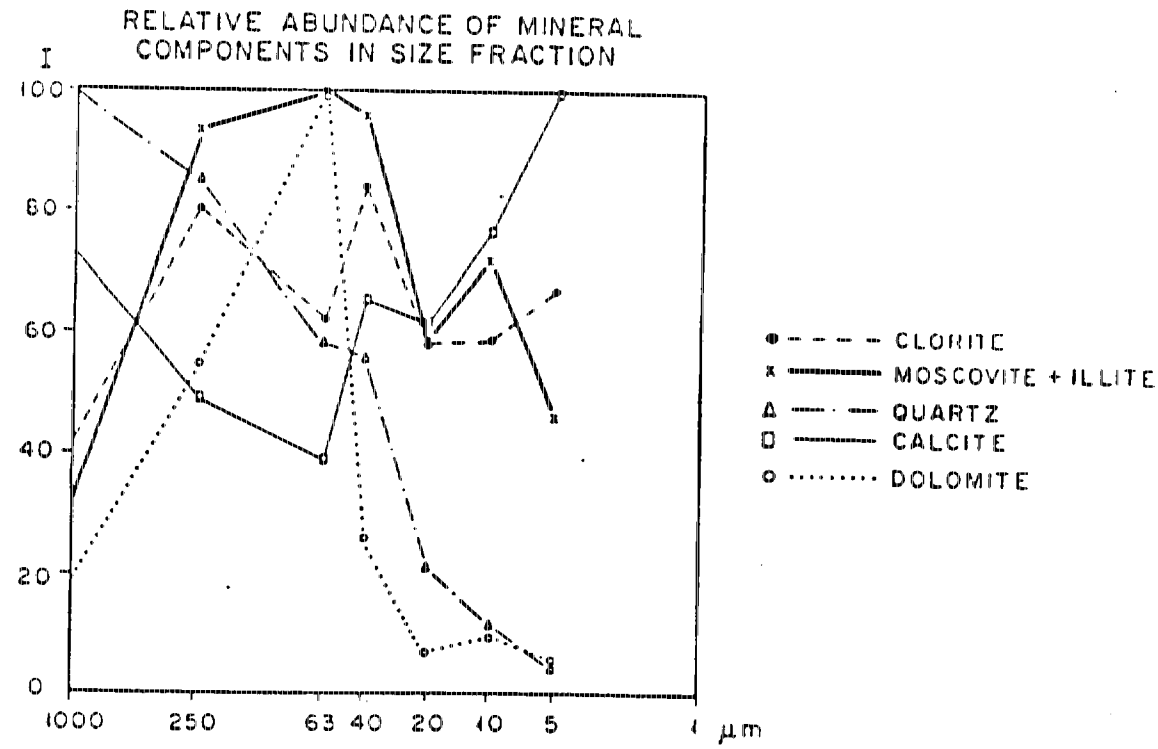


FIG. - 3

PARCEL 2 - O



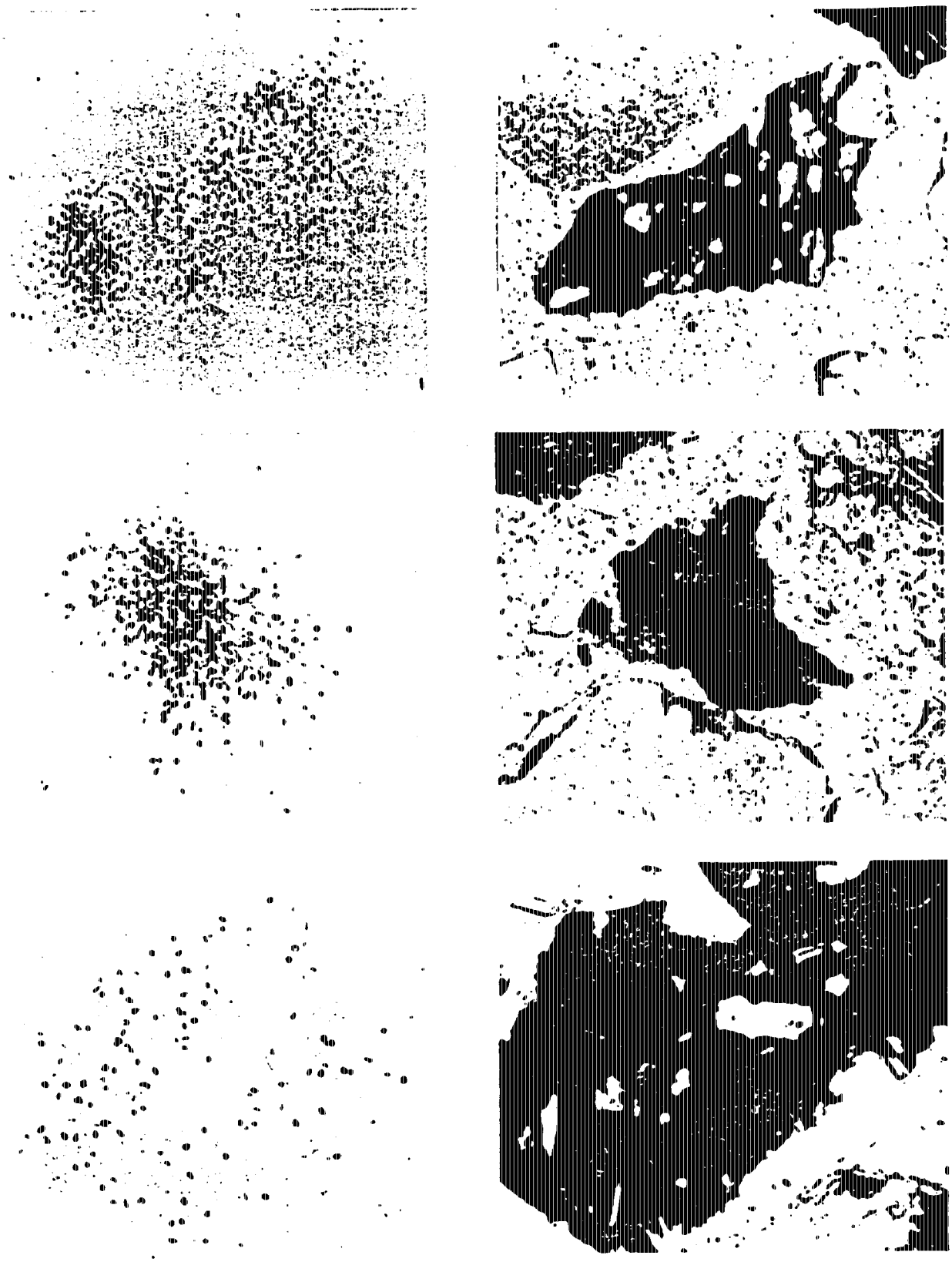


Fig.4. ⇐ Autoradiography of iron oxy-hydroxides. L.N. x 75 and 150

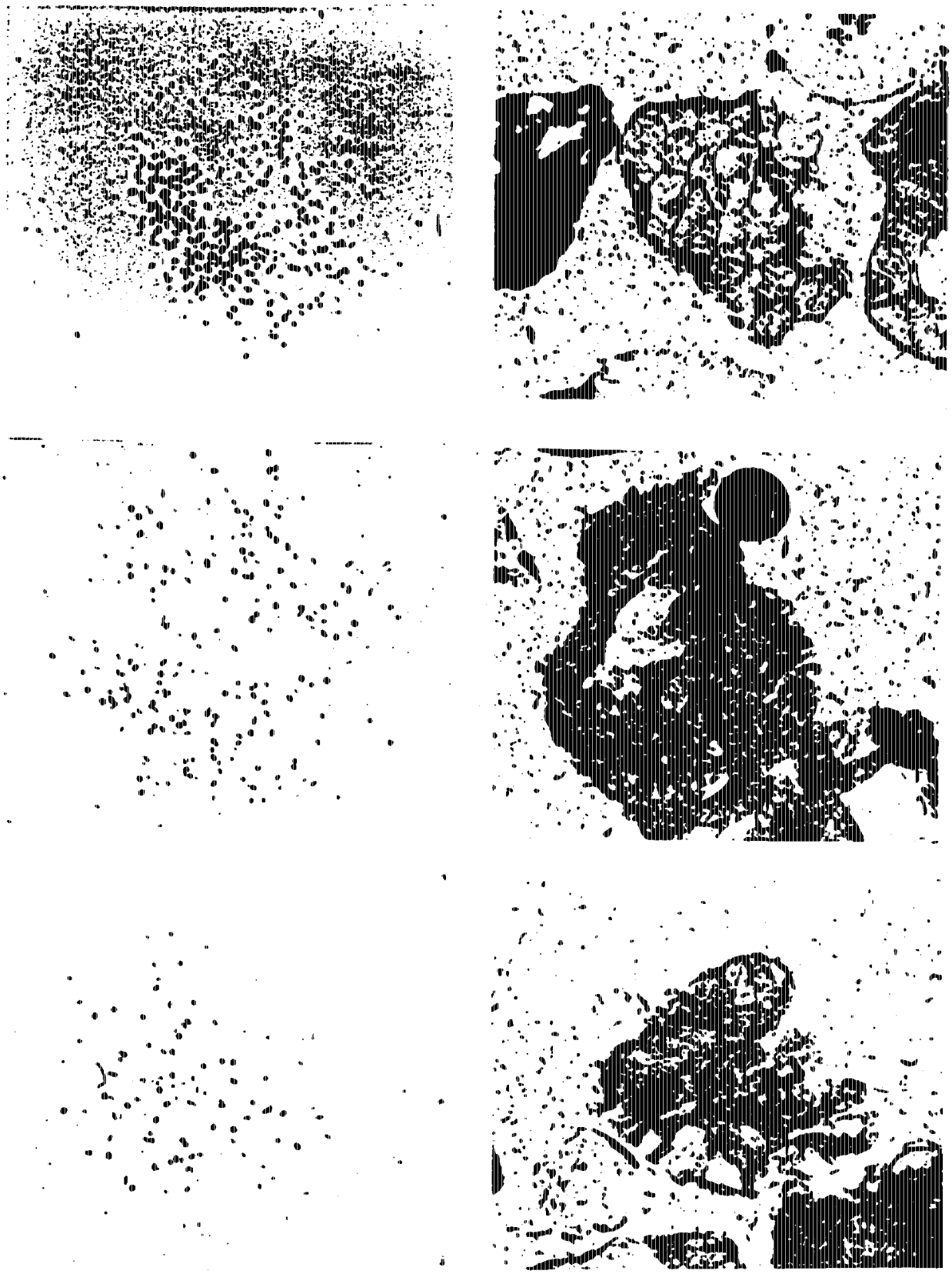


Fig.5. ⇐ Autoradiography of carbonates and micaceous aggregates. L.N.x150

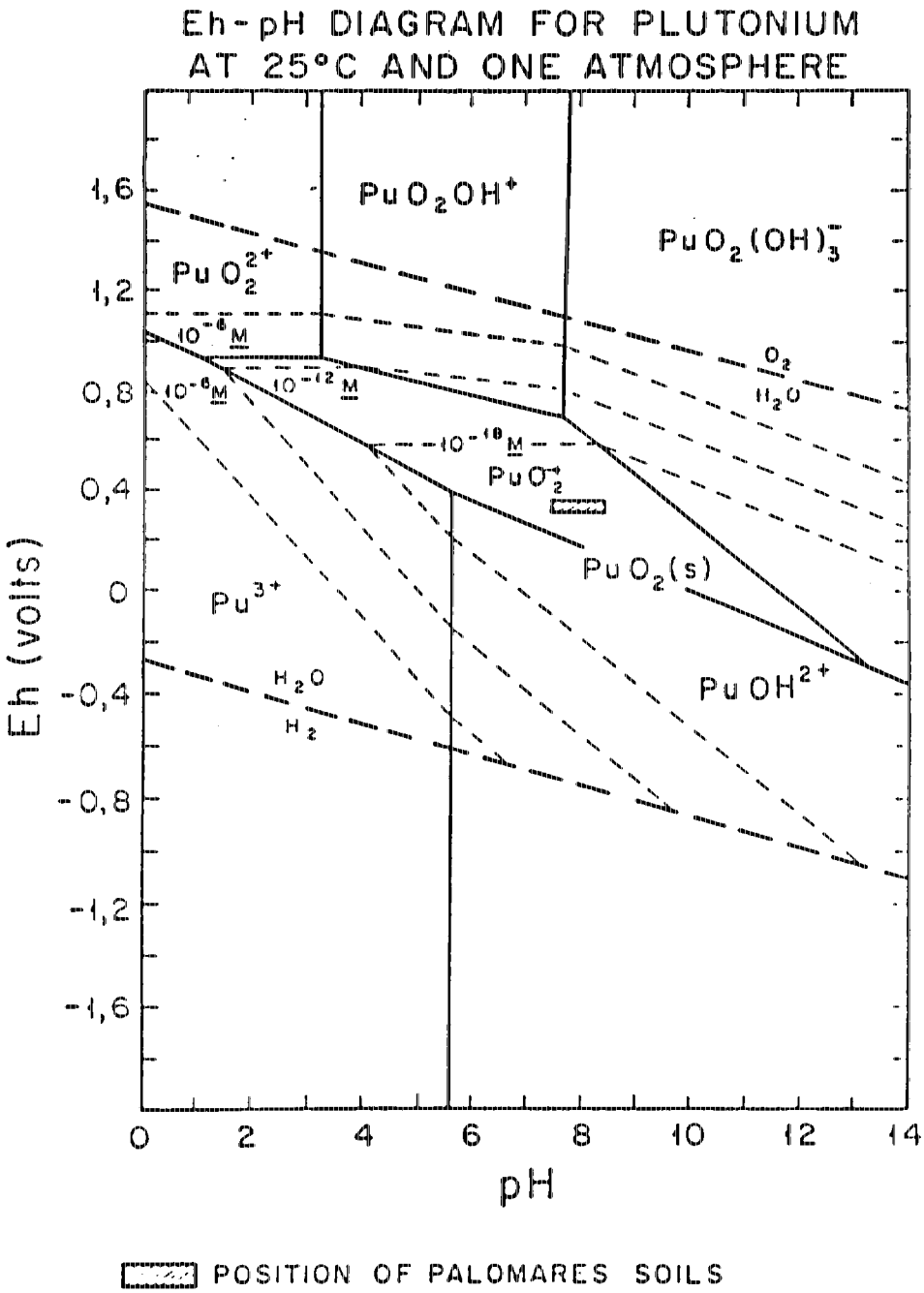

Eh - pH DIAGRAM FOR PLUTONIUM
AT 25°C AND ONE ATMOSPHERE
PuO2OH+
PuO2(OH)3-
PuO2 2+
10-6 M
10-6 M
10-12 M
10-18 M
PuO2+
PuO2(s)
Pu3+
PuOH2+
H2O
H2
O2
H2O
Eh (volts)
1,6
1,2
0,8
0,4
0
-0,4
-0,8
-1,2
-1,6
0
2
4
6
8
10
12
14
pH
POSITION OF PALOMARES SOILS

FIG.- 6